METROLOGY OF RADIONUCLIDES

PROCEEDINGS

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INTERNATIONAL ATOMIC ENERGY AGENCY
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METROLOGY OF RADIONUCLIDES
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The Agency’s Statute was approved on 26 October 1956 at an international conference held at United Nations headquarters, New York, and the Agency came into being when the Statute entered into force on 29 July 1957. The first session of the General Conference was held in Vienna, Austria, the permanent seat of the Agency, in October, 1957.

The main objective of the Agency is “to accelerate and enlarge the contribution of atomic energy to peace, health and prosperity throughout the world”.

Printed in Austria
"Metrology of Radionuclides" is the science of precise measurements of the absolute value of the activity of radioactive sources. A rapid expansion has taken place over the past few years in the applications of radionuclides in various fields of scientific research, particularly in the production of commodities which lead to improved living standards. This has occurred not only in the countries most advanced in nuclear science, but in many others.

In order to allow those actively engaged in this field to exchange research results and discuss their problems, the International Atomic Energy Agency sponsored a symposium which was held in Vienna from 14—16 October, 1959. Thirty-seven papers were presented from 14 countries. These covered a general survey on the routine methods of standardization of radionuclides and new developments of absolute measuring methods for their standardization.

The results of these proceedings are now published by the Agency in the present volume, in accordance with its Statute to foster the exchange of scientific and technical information on the peaceful uses of atomic energy. It is hoped that it will prove useful to everyone interested in the subject, and of particular value to scientists and institutions entering this field.

June 1960

[Signature]

Director General
CONTENTS

INTRODUCTION .................. XI

I. GENERAL SURVEY PAPERS ON THE ROUTINE METHODS OF
STANDARDIZATION OF RADIONUCLIDES

I. 1. P. J. Campion, K. W. Geiger
"Routine methods of standardization in Canada" .................. 3

I. 2. B. Grinberg
"L'Étalonnage des radioéléments en France" .................. 21

I. 3. Masashi Naito, Yajiro Inouye, Osamu Yura
"Standardization of radionuclides in the Electrotechnical Laboratory, Tokyo" .................. 33

I. 4. B. M. W. Robinson
"Routine standardization of radionuclides in the UK" .................. 41

I. 5. H. M. Weiss
"Absolute measurement of radioactive materials at the Physikalisch-Technische
Bundesanstalt" .................. 53

I. 6. K. K. Aglintsev, V. V. Bochkarev, V. N. Grablewskii, F. M. Karavaev
"Metody metrologii radioaktivnosti v SSSR" .................. 61

I. 7. W. B. Mann
"Routine methods of radioactivity standardization at the National Bureau of
Standards (USA)" .................. 89

I. 8. G. G. Manov
"Techniques of large scale processing of counting data" .................. 103

I. 9. L. M. Cavallo and W. B. Mann
"The organization of international intercomparisons of radioactivity standards
with special reference to such measurements of NBS standards" .................. 117

I. 10. J. Braun
"Standardization of radionuclides at the Atomic Energy Company (Sweden)" .................. 123

Discussion

II. NEW DEVELOPMENTS OF ABSOLUTE MEASURING METHODS
FOR THE STANDARDIZATION OF RADIONUCLIDES

II. 1. α-EMITTING NUCLIDES .................. 147

II. 1. 1. H. P. Robinson
"A new design for a high precision, high geometry alpha counter" .................. 147

II. 1. 2. J. Steyn and F. W. E. Strelow
"The determination of the half-life of U\textsuperscript{238} by absolute counting of alpha
particles in a 4π liquid scintillation counter" .................. 155

II. 2. β-EMITTING NUCLIDES .................. 163

II. 2. 1. B. D. Pate
"Disintegration-rate determination by 4π counting" .................. 163

II. 2. 1. N. G. Trott, R. E. Bentley and L. K. Burton
"Demountable 4π Geiger counters for standardization and the assay of low-
activity samples" .................. 175

II. 2. 2. L. Yaffe and J. B. Fishman
"Self-absorption studies with a 4π β-proportional flow counter" .................. 185
II. 2. 3. A. M. Baptista
"On absolute measurements of β-emitting radionuclides" 201

II. 2. 4. P. J. Campion, J. G. V. Taylor and J. S. Merritt
"The absolute counting of nuclides decaying by β emission only" 227

II. 2. 5. Y. Le Gallic, B. Grinberg, M. Thenard
"Remarques sur une technique rapide de mesure et d'analyse d'émiteurs β en solution de très faible activité spécifique" 245

II. 2. 6. G. G. Manov
"Low-level β and γ counting in the region 0—10 disintegrations per minute" 253

II. 2. 7. M. Forte, A. Anzani
"A method for scintillation counting of very-low-energy particles (below 1 keV)" 269

II. 2. 8. J. Steyn
"The comparative accuracy of the 4π liquid scintillation counting method of radioisotope standardization" 279

II. 2. 9. J. J. Engelmann
"Mesure des émetteurs β par la charge rayonnée" 291

II. 2. 10. K. Hogrebe
"The absolute measurement of the radioactivity of gases" 301

II. 2. 11. W. B. Mann
"Recent work with compensated internal gas counters for the standardization of gaseous radionuclides" 307

II. 2. 12. V. V. Bochkarev, V. A. Bajenov
"О некоторых методах измерения β-активных газов" 317

II. 2. 13. R. V. Gasström
"On the Single-Scintillator Coincidence Technique" 333

Discussion

II. 3. ELECTRON-CAPTURE NUCLIDES 343

II. 3. 1. R. A. Allen
"Some remarks on the progress in electron capture standardization" 343

II. 3. 2. S. C. Curran
"The proportional counter — some aspects of operation" 353

Discussion

II. 4. GAMMA COUNTING 375

II. 4. 1. K. Waechter
"The proof sensitivity of well-type crystals of different sizes with regard to quantum-radiation from common radionuclides" 375

Discussion

III. NEUTRON SOURCE CALIBRATION 387

III. 1. J. Thomas, P. Skjerk Christensen, Bent Fastrup, J. Olsen, P. Villemoes
"Low level standardization at Risø" 387

III. 2. K. W. Geiger
"Absolute β counting and its application for the standardization of radioactive neutron sources" 393

Discussion

IV. HIGH INTENSITY SOURCE MEASUREMENTS 405

IV. 1. J. Domanus, B. Osuchowski
"The concept of 'gramme-equivalent-of-radium' for non-point isotopic γ-ray sources" 405
IV. 2. B. Gross
“Batteryless γ-ray dosimeter” 413

IV. 3. A. Brynjolfsson, N. W. Holm
“Calorimetric measurements of γ-rays and calibration of ferrous sulphate radiation dosimeter” 419

Discussion

V. MICROCALORIMETRIC METHODS OF STANDARDIZATION

V. 1. M. Lecoin
“La méthode de microcalorimétrie adiabatique à compensation automatique. — Application à la comparaison des étalons de radium” 329

Discussion

VI. PHOTOMULTIPLIERS

VI. 1. G. Kainz, K. Schärf, R. Patzelt
“On the mechanism of fatigue effects in photomultipliers” 439

VII. APPLICATION OF ABSOLUTE MEASUREMENT METHODS TO VARIOUS PROBLEMS

VII. 1. T. B. Novey
“Methods of particle detection in free neutron decay” 449

VII. 3. J. L. Putman
“Estimation of a distributed β-emitter by measurements of bremsstrahlung” 459

Discussion

Conclusions

LIST OF PARTICIPANTS 469
INTRODUCTION

Mr. H. de LABOULAYE, Acting Director General, opened the inaugural session of the Symposium with the following speech:

Poursuivant la réalisation de ses objectifs statutaires en matière d’information scientifique et technique, l’Agence internationale de l’énergie atomique a organisé le présent Colloque sur “la métrologie des radionuclides”. J’ai l’honneur d’ouvrir ce colloque au nom du Directeur général, Mr. Sterling Cole, qui regrette de se voir empêché — étant absent de Vienne — de présider cette séance inaugurale. Mr. Sterling Cole m’a chargé de vous transmettre ses salutations et ses voeux pour la réussite de vos travaux.

Au cours de ces dernières années, l’utilisation des radionuclides est allée en augmentant très rapidement, aussi bien du point de vue du nombre des applications pacifiques dans la recherche scientifique et dans la production des biens destinés à accroître le bien-être et le niveau de vie des peuples, que du point de vue de la variété et des quantités de radionuclides mises en jeu. D’autre part, l’usage des radionuclides, encore limité — jusqu’à un passé récent — à un nombre restreint de pays avancés en matière de science et de technique nucléaires, commence à se répandre de plus en plus rapidement dans un nombre croissant d’autres pays. Cette évolution — dont on peut facilement prédire qu’elle continuera dans le même sens à l’avenir — confère aux problèmes de la métrologie des radionuclides l’importance pratique de premier plan, dont vous êtes, en tant que spécialistes, parfaitement conscients.

Le but de la métrologie des radionuclides est d’élaborer des méthodes de mesure capables de fournir, avec une précision aussi grande que possible, la valeur absolue de l’activité des sources utilisées. Le problème est complexe, étant donné qu’il implique des recherches dans différents domaines, comme par exemple la mise au point de détecteurs de rayonnements aussi efficaces que possible et de circuits électroniques à caractéristiques de plus en plus poussées, la connaissance de plus en plus exacte des vies moyennes des radionuclides et de leurs schémas de désintégration, la mise au point de méthodes commodes, rapides et précises de détermination de la composition isotopique des substances actives constituant les sources, etc.

Au cours des dernières années, des efforts considérables et souvent couronnés d’un remarquable succès ont été faits dans ces diverses directions. Il a paru au Comité scientifique consultatif de notre Agence qui, comme vous le savez, est composé de quelques-unes des plus éminentes personnalités des sciences nucléaires, qu’il serait important d’essayer d’opérer une synthèse, sur le plan mondial, de l’expérience acquise dans le domaine de l’étalonnage des radionuclides. C’est le but que se propose d’atteindre le programme de la première journée du Colloque, qui est consacrée précisément à l’étude comparative des méthodes courantes d’étalonnage, actuellement utilisées dans les laboratoires spécialisés de différents pays.

Mais il est certainement plus important encore, d’essayer de dégager les tendances qui se manifestent à l’heure actuelle dans le développement et le raffinement des méthodes de mesure de la radioactivité. C’est le but que poursuit le programme des deux journées suivantes de notre réunion.
Les prévisions les plus optimistes du Secrétariat, en ce qui concerne l’intérêt soulevé par le présent Colloque, ont été largement dépassées. Cet intérêt est témoigné par le nombre considérable des mémoires présentés: le Secrétariat éprouve même la crainte que ce nombre ne soit trop élevé pour la durée du Colloque et qu’il ne vous impose un rythme de travail assez fatigant. Si tel est le cas, nous nous en excusons à l’avance. Nous nous sommes efforcés de réserver aux discussions le maximum de temps possible, car il nous semble — et nous pensons que vous partagez ce sentiment — que ce sont précisément les discussions — à condition qu’elles soient animées — qui constituent la partie la plus vivante et la plus fructueuse d’une conférence scientifique.

L’intérêt soulevé par notre Colloque est également illustré par le grand nombre d’experts et de participants réunis dans cette salle et qui proviennent d’un grand nombre d’États-Membres. J’exprime le vœu que le déroulement des travaux du Colloque couronne les efforts déployés pour le préparer. A ce sujet, le Secrétariat tient à remercier chaleureusement tous ceux qui l’ont appuyé dans son travail: les auteurs des communications ainsi que les membres du Comité des étaisons et mesures de radioactivité à usage radiologique de la Commission internationale des unités et des mesures radiologiques.

Les actes du Colloque seront publiés par les soins de l’Agence et contiendront le texte intégral des mémoires ainsi que le compte-rendu des discussions engagées en marge des exposés. Nous espérons que ce recueil d’information scientifique sera d’une réelle utilité, surtout pour les États-Membres qui sont engagés depuis peu dans le domaine de la recherche et des applications pacifiques de l’énergie nucléaire, ou qui s’y engageront dans un avenir rapproché. L’Agence internationale de l’énergie atomique pourrait alors estimer qu’elle a atteint, dans un de ses champs d’activité, les buts pour lesquels elle a été créée.

Avant de céder la place au Président de la première séance, vous me permettrez ici d’exprimer notre émotion de ne pas voir dans cette salle deux de vos collègues dont la participation à ce colloque était prévue. Il s’agit du Dr. W. E. Perry du Royaume-Uni, et du Dr. Gerhard Kainz, d’Autriche, décédés, le premier il y a quelques mois, le second il y a quelques jours à peine. Je suis sûr que vous tiendrez à vous incliner comme moi devant leur mémoire.

Pour terminer, ce m’est un plaisir de céder la Présidence de cette première séance à Madame le Professeur Berta Karlik, qui dirige l’Institut du Radium et de Recherches Nucléaires de l’Académie des Sciences Autrichienne. Cet Institut est, si je ne me trompe, le plus ancien du genre au monde.

Madame Karlik est bien connue de vous tous par ses travaux comme l’une des figures les plus éminentes de la recherche nucléaire en Autriche.
I.

GENERAL SURVEY PAPERS ON THE ROUTINE METHODS OF STANDARDIZATION OF RADIONUCLIDES

(SESSIONS 1 AND 2, 14 OCTOBER 1959)
I. 1. ROUTINE METHODS OF STANDARDIZATION IN CANADA

P. J. CAMPION AND K. W. GEIGER

Abstract

The standardization of β-active nuclides is undertaken primarily by Atomic Energy of Canada Limited using the $4\pi$ β and $4\pi$ β-γ coincidence techniques. The detectors used are gas flow counters operated in the proportional region.

For gaseous activities the method of differential gas counting is employed while for those nuclides which decay by electron capture a high pressure $4\pi$ proportional counter is available.

Radium standardization, by ionization measurement, is carried out at the National Research Council, Ottawa. The ionization chamber and associated circuitry will be described; an accuracy for intercomparison of sources of 0.1% may be achieved. The same ionization chamber can also be used to calibrate Co$^{60}$ sources in millicurie range; the radium and cobalt sources calibrated routinely are mainly for medical application. Standardization of other nuclides has been carried out by determining their specific γ-ray emission.

The absolute calibration of neutron sources is also undertaken using various methods, while intercomparison of neutron sources is effected by long BF$_3$ counters.

Recently, a $4\pi$ β-γ coincidence apparatus has been built which basically follows the design of Atomic Energy of Canada Limited.

Méthodes courantes d'étalonnage des radionuclides au Canada

Les émetteurs β sont étalonnés surtout par l'Atomic Energy of Canada Limited, qui utilise des dispositifs $4\pi$ β et $4\pi$ β-γ à coïncidences. Les appareils sont des compteurs à gaz travaillant dans la région dite "proportionnelle".

Pour les gaz radioactifs, on utilise la méthode différentielle de comptage et pour les nucléides à capture électronique, il existe un compteur proportionnel $4\pi$ à haute pression.

L'étalonnage du radium par mesure de l'ionisation se fait au National Research Council, à Ottawa. Les auteurs décrivent la chambre d'ionisation et les circuits associés; il est possible de comparer les sources entre elles avec une précision de 0.1%. La chambre d'ionisation en question peut également servir à l'étalonnage de sources de Co$^{60}$ d'une activité de l'ordre du millicurie; les sources de radium et de cobalt ainsi étalonnées sont destinées surtout à des applications médicales. On a étalonné d'autres nucléides en mesurant leur émission spécifique des rayons γ.

L'étalonnage absolu des sources de neutrons se fait par plusieurs méthodes et l'on compare ces sources entre elles à l'aide de compteurs longs au trifluorure de bore.

Un dispositif à coïncidences $4\pi$ β-γ a été récemment construit d'après les plans de l'Atomic Energy of Canada, Limited.
Обычные методы стандартизации, применяемые в Канаде

Стандартизация β-активных изотопов предпринимается в первую очередь компанией Атомик Энерджи оф Канада Лимитид, использующей 4 π β и 4 π β-γ счетчики совпадений. В качестве детекторов использовались пропорциональные газовые счетчики.

Для определения газовых активностей применяется метод дифференциального газового исчисления, для тех же изотопов, которые распадаются при захвате электронов, имеется пропорциональный счетчик 4 π высокого давления.

Стандартизация радия при помощи измерения ионизации осуществляется Национальным научно-исследовательским Советом в Оттаве. Будут описаны ионизационная камера и связанные с ней схемы. При сравнении источников может быть достигнута точность в 0,1‰.

Та же самая ионизационная камера может быть также использована для калибровки источников Co⁶⁰ в пределах милликрюра; источники радия и кобальта, калиброванные обычным способом, применяются, главным образом, для медицинских целей. Стандартизация других изотопов была осуществлена посредством определения их удельного γ-излучения.

Абсолютная калибровка источников нейтронов осуществляется также и с помощью различных методом, а их сравнение между собой длинными BF₃ счетчиками.

Недавно был создан 4 π β-γ счетчик совпадений. Он в основном совпадает с конструкцией компании Атомик Энерджи оф Канада Лимитид.

Métodos corrientes de normalización de radionúclidos en el Canadá

La entidad que se ocupa principalmente de la normalización de los radionúclidos β es la Atomic Energy of Canada Limited, que emplea para ello métodos β 4 π y β-γ 4 π de coincidencias. Como detectores se utilizan contadores de corriente gaseosa con los que se trabaja en la zona de proporcionalidad.

Para determinar la actividad de sustancias gaseosas se recurre al método diferencial de recuento, en tanto que para los núclidos que se desintegran por captura electrónica se emplea un contador proporcional 4 π de elevada presión.

La normalización del radio por medida de la ionización se efectúa en el National Research Council, Ottawa. Los autores describen la cámara de ionización y los circuitos auxiliares; en la comparación de las fuentes puede alcanzarse una precisión del 0.1‰. La misma cámara de ionización puede también servir para calibrar fuentes de Co⁶⁰ cuya actividad sea del orden de los milicuries; las fuentes de radio y cobalto así calibradas se destinan por lo general a aplicaciones médicas. Se ha efectuado la normalización de otros radionúclidos determinando la radiación γ específica que emiten.

Asimismo, se efectúa la calibración absoluta de fuentes neutrónicas siguiendo diferentes métodos; la comparación de dichas fuentes se hace con ayuda de contadores largos de BF₃.

Recentemente, se ha costruito un contador de coincidencias β-γ 4 π, que, en líneas generales, presenta características análogas a las del aparato de la Atomic Energy of Canada Limited.
Two government institutions in Canada are actively engaged in radio-nuclide standardization. The responsibility for fundamental standards is vested in the National Research Council in Ottawa, and Dr. Geiger will describe the work which is done there. The other institution is Atomic Energy of Canada Limited, Chalk River (some 130 miles north-west of Ottawa). Because of the importance of β-ray standardization to nuclear physics, reactor technology, and the production of radionuclides we have been more actively concerned with this aspect of standardization and I will now endeavour to present some of the routine techniques which are used at Chalk River.

For many years $4\pi$ β counting using gas flow counters in the proportional region has been the preferred routine method for β emitters having end point energies greater than about 300 keV. The $4\pi$ counters are quite simple, and a photograph of one is shown in figure 1. This is a pill box type with a 0.013 mm wire in both halves. A source mount ring with a thin plastic
film stretched across it is also shown in figure 1 together with a convenient storage container for the source and mount. The counting rate is measured as a function of the voltage applied to the counter anode. This yields the well known plateau region which for our particular counter-amplifier systems is about 200 to 300 volts long and has a slope of about one to two tenths of one per cent per 100 volts. Methane is normally used for the flow gas, although a mixture consisting of 90% argon and 10% methane is used for special purposes. Other types of counters such as the spherical type with loop anodes have been used with similar results.

Source preparations and all bulk handling of radioactive materials are carried out in a chemical laboratory well separated from the counting room. Sources are deposited volumetrically on thin plastic films of VYNS of about 5 to 10 μg/cm². These are coated on the underside with about 10—15 μg/cm² of gold to render them conducting. A source is deposited on the top surface and when dry is covered with another similar film so that both gold surfaces are outermost. The second film adheres well to the first and to the source material without forming cavities. For most routine measurements the thickness of these films can be adequately judged by interference colours. For certain applications, however, a more precise value of the thickness is sometimes required and to obtain this we have designed a small β-ray thickness gauge which is shown in figure 2. The gauge consists of a 2 π counter having a thin VYNS window mounted on a supporting mesh. On the underside of the counter, on a hydraulically operated platform, is a thick tritium source. Methane is passed through the counter while hydrogen is passed through the space between the window and tritium source. The transmission of the β-rays from this source through a film under test is measured and the thickness in μg/cm² determined from a
calibration curve which was obtained by weighing sample films on a quartz fibre microbalance. The sensitivity of the gauge is about 1.5% per microgram.

Recently the merits of several source preparation techniques have been investigated (1). Besides the normal evaporation of an aliquot under a heat lamp there are two others which are convenient to use for routine work. The first, the use of insulin as a spreading agent, is probably well known to all present. The second is the use of colloidal silica (1). This technique is thought to be a seeding process and was suggested by Mr. W. F. Merritt, of the Chalk River Laboratories. A few microlitres of a very dilute aqueous colloidal silica solution are added to the aliquot. The silica particles are, according to the manufacturer’s data, quite uniform in size and only 60 angstroms in diameter and act as seeds for the crystallization process. Since these ‘seeds’ are uniformly distributed throughout the aliquot the resulting deposit consists of a large number of evenly spaced small crystals rather than a few large ones.

The recording circuit of our 4πβ counting system consists of an amplifier and scaler together with an interval timer controlled by a frequency standard. The dead time of the system is determined by the amplifier and is about 2 μsec. It is thus possible to work with sources of a few thousand counts per second and yet keep both the dead time and background corrections small. The dead time is measured using the paired source technique and a photograph of these paired sources is shown in figure 3. The disc, which has the same diameter as our standard source mount rings shown earlier, has two circular holes in which sources of Sr90—Y90 sandwiched between thin aluminum plates may be placed. Blanc source mounts are available for single source and background measurements.

Figure 3
Paired sources and mount for dead time measurements
In addition corrections are also required for source mount and film absorptions, the greater being that due to self-absorption. For routine measurements an estimate of the self-absorption is made from an empirical curve relating this quantity to the β end point energy (1). This curve was obtained by the direct measurement of the self-absorption of a number of nuclides which are amenable to the $4\pi\beta-\gamma$ coincidence technique and is discussed in more detail in another paper.* The film correction is determined by applying pairs of additional films to the source mount and making the extrapolation to zero film thickness. Typically this correction amounts to 1% for Co$^{60}$ and about 0.05% thickness. Typically this correction amounts to 1% for Co$^{60}$ and about 0.05% for Na$^{24}$.

Because of the large uncertainty in the self-absorption in β emitting sources the use of the $4\pi\beta-\gamma$ coincidence method (2) is preferred whenever possible. Figure 4 shows the coincidence unit which consists of a $4\pi$ proportional flow counter sandwiched between two $3''\times3''$ NaI crystals. The shielding consists of 4'' to 6'' of lead and part of this may be rolled back along the supporting table to allow easy access to the $4\pi$ counter. The electronics

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* Paper II, 2. 4., page 227.
associated with this unit are contained in the two adjacent racks. The $4\pi$ counter itself is shown in an exploded view in figure 5. It is essentially a pill-box type with lucite inserts to avoid regions of reduced electric field strengths. Loading the unit is effected by placing the source mount on the turntable which is then mechanically rotated to bring the source into the centre of the counter.

![Diagram of coincidence counters]

**Figure 5**

Details of coincidence counters

The use of two NaI crystals makes for greater flexibility of the instrument. For example, it is possible to do triple coincidences, or alternatively one may add the output signals from both detectors linearly in order to increase the effective size of the $\gamma$-ray detector. This is useful when looking at very weak $\gamma$-ray branches or when one wishes to accept pulses in the $\gamma$ channel which correspond to a sum peak. Normally, for sources such as $^{60}\text{Co}$ for example, the efficiency of the $\gamma$ detectors is about 10 to 20% depending on the gate settings of the pulse-height analysers. Thus a measurement of reasonable statistical accuracy can be made in about 3 or 4 minutes. In this respect the statistics in the $4\pi \beta-\gamma$ coincidence method are better than those given by the total counts accumulated in the coincidence scaler. To demonstrate this let us assume an ideal case in which the $4\pi \beta$ counter-source combination is exactly 100% efficient and there is no background rate in either detector. Then for every count recorded in the $\gamma$ detector there would be a count in the coincidence channel, and thus the total counts recorded in this channel are an exact fraction of those recorded in the $\beta$ detector. Thus in this case the statistics are determined by the accumulated counts in the $\beta$ scaler. The introduction of efficiencies less than 100% obviously modifies this in any
practical case. The equation relating the fractional standard deviation \( \sigma \) in the absolute disintegration rate to the number of true counts accumulated in the coincidence channel, \( n_c \), and the efficiencies of the detectors \( (\varepsilon_\beta, \varepsilon_\gamma) \) is:

\[
\sigma = \left[ \frac{1}{n_c} \left( 2\varepsilon_\beta \varepsilon_\gamma - \varepsilon_\beta - \varepsilon_\gamma + 1 \right) \right]^{1/2}.
\]

For very small efficiencies this reduces to \( 1/\sqrt{n_c} \) as expected. The variation of this statistical error is shown graphically in figure 6, where in order to demonstrate this variation in a way which is independent of any counting rate, \( \sigma \sqrt{n_c} \) rather than \( \sigma \), has been plotted against the \( \beta \) detector efficiency, and curves are shown for four different efficiencies of the \( \gamma \) detector.

![Figure 6](image)

Variation of \( \sigma \sqrt{n_c} \) as a function of \( \varepsilon_\beta \)

It is felt that the \( 4\pi \beta-\gamma \) coincidence method is capable of quite high accuracy and hence of more consistent results. This consistency is demonstrated by the results of a recent distribution of a solution of Au\(^{198}\). A sufficient number of answers were obtained by \( 4\pi \beta-\gamma \) coincidence counting to make an interesting comparison between these results and those obtained by straight \( 4\pi \) counting in the same distribution. The distribution was made by the National Bureau of Standards in November 1958, and these results are made available through courtesy of the Bureau and the individual laboratories taking part. Laboratories were requested to report the activity measured on three consecutive days and the results were corrected for decay to a reference time on the central day. Since in this analysis we wish to compare techniques we have averaged the results obtained on the three days by any one laboratory for any one technique. This eliminates any undue weight being given to a determination for which results for all three days may have been quoted as against another determination for which results for only one day were quoted. The results of this analysis are given in Table I.
TABLE I

Au¹⁰⁸ Intercomparison; November 1958

<table>
<thead>
<tr>
<th>Laboratory</th>
<th>4πβ</th>
<th>4πβ-γ</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atomic Energy of Canada Limited</td>
<td>—</td>
<td>1.331</td>
</tr>
<tr>
<td>Commissariat à l’Energie Atomique</td>
<td>1.350</td>
<td>—</td>
</tr>
<tr>
<td>Fondation Curie</td>
<td>1.307</td>
<td>—</td>
</tr>
<tr>
<td>Kernreaktor, Bau- und Betriebs-Gesellschaft m. b. H.</td>
<td>1.320</td>
<td>—</td>
</tr>
<tr>
<td>National Bureau of Standards</td>
<td>1.297</td>
<td>—</td>
</tr>
<tr>
<td>National Physical Laboratory</td>
<td>1.313</td>
<td>—</td>
</tr>
<tr>
<td>National Physical Research Laboratory</td>
<td>1.287</td>
<td>—</td>
</tr>
<tr>
<td>National Physical Research Laboratory</td>
<td>1.302</td>
<td>—</td>
</tr>
<tr>
<td>National Research Council</td>
<td>1.321</td>
<td>1.324</td>
</tr>
<tr>
<td>Physikalisch-Technische Bundesanstalt</td>
<td>1.311</td>
<td>1.339</td>
</tr>
<tr>
<td></td>
<td>1.288</td>
<td>1.328</td>
</tr>
<tr>
<td>Mean Values</td>
<td>1.310 ± 0.018</td>
<td>1.330 ± 0.005</td>
</tr>
</tbody>
</table>

Statistical weighting of these results is hardly justified and in obtaining the mean values shown in Table I equal weight has been given to each result. The errors associated with these means are the root mean square deviations. The deviation in the 4πβ-γ results is less than four tenths of one per cent which is remarkably good agreement between four independent laboratories. This is almost four times smaller than that associated with straight 4πβ counting and is largely due to the elimination of self-absorption in the coincidence method. Of course there may be other systematic errors to which the 4πβ-γ coincidence method is susceptible but on the basis of these results such errors are unlikely to be very large. One such possible error is the assumption of 100% coincidence mixer efficiency. The technique of determining the coincidence counting rate as a function of the relative delay between channels for different resolving times is a useful method for checking this. Further, while efficiencies approaching very close to 100% have been observed we have not measured an apparent efficiency of greater than 100%. The highest efficiency observed to date is 99.9% with a liberal error of 0.2% and was observed for the positron branch of Cu⁶⁴ (1). If it is assumed that the coincidence mixer efficiency cannot be greater than 100% then such a measurement implies that this efficiency is unity to within 0.1 or 0.2%.

For the rare gas activities and those nuclides which can be readily converted into a gaseous phase we have a system of differential gas counters. These consist of pairs of cylindrical proportional counters, the members of each pair being made similar in all respects except in the length of the active volume. The two counters are filled to the same pressure of active gas as measured by a cathetometer. This pressure is usually a few centimetres of Hg and is made up to a total of about 60 or 70 cms of Hg by the addition of inactive methane. In common with other laboratories it is found necessary to mechanically mix the two gases, since straightforward diffusion seems to take far too long. The counters are removed from the gas filling line and counted behind lead shielding. The slope of the plateau of counts per unit time against applied voltage is about 0.5% per 100 volts. However the

11
difference in counting rate between the two counters at any given voltage yields a line whose slope is about 0.1% per 100 volts, this reduction being due to the cancellation of end effects in the counters. Dr. W. B. Mann has described more fully a similar technique which is used at the National Bureau of Standards.

In the case of electron capturing nuclides there are two methods used for standardization. The first, the coincidence method, may be used for those nuclides which have a γ-ray in their decay scheme. We use the coincidence equipment shown in figure 4, but in order to increase the efficiency of the 4π counter, use is made of a pre-mixed flow gas consisting of 90% argon and 10% methane. The other method uses a 4π high pressure counter. Because self-absorption effects are probably rather large for the Auger electrons we prefer to sandwich the activity between foils of sufficient thickness to stop these electrons entering the gas of the 4π counter. The X-ray emission rate is then determined by increasing the pressure until essentially all the X-rays are stopped within the gas of the counter. Any further increase in pressure does not yield an increased counting rate and this is the so-called 'pressure' plateau (3). To obtain the X-ray emission rate from the source a correction must be made for the absorption of the X-rays in the sandwiching foils and this is accomplished by means of the well known Gold Integral. The accuracy of this method is limited by the uniformity of the sandwiching foils and the knowledge of the fluorescence yield of the product nucleus. A combination of these two methods has recently been used to determine the fluorescence yield of vanadium and chromium (4).

One further instrument that is in routine use at Chalk River is a 4π ionization chamber filled to 20 atmospheres of argon for γ-ray measurements. In itself this is not an absolute instrument but in conjunction with a reference source of radium and a previous calibration with the nuclide concerned it forms a very useful means of secondary standardization. It also provides a very quick method for relative measurements.

REFERENCES


Under the act of its institution the National Research Council has the responsibility for fundamental physical standards. This includes radionuclide standardization and official certificates are issued for such work.

Radium standardization at the National Research Council dates back thirty years and a few words about this subject may be in place although many people think this field is of little importance today. Radium standardization is quite different from the type of standardization described just now by Dr. Campion. The difference arises out of the fact that Ra\(^{226}\) is one of the few radioactive nuclides the amount of which can be determined by weighing. On the other hand, because of the many radioactive decay products present with radium, absolute counting is a rather difficult task. Because of the high cost of radium and because of its widespread use in the medical field, radium standardization became important soon after its discovery.

In the nineteen twenties Canada was one of the important radium producing countries and in 1930, two years after the National Research Council opened its first laboratories in Ottawa, equipment for radium standardization was set up. Since a weighed radium standard was not available, standardization was based on standards the content of which was...
not known by weighing but rather by \( \gamma \) intercomparison at laboratories of other countries. It was only in 1955 that a weighed standard was acquired; this was one out of the twenty standards Prof. Hönigschmid prepared in 1934. It seems that nobody has prepared weighed standards since that time which may indicate the difficulties in their preparation and perhaps also the diminishing interest in radium standardization.

Figures 1 and 2 show the apparatus which is used at the National Research Council for comparison of radium sources and which had existed in its basic form for many years. The radium source is placed in a V-shaped hod, at a certain distance from an ionization chamber. This is a parallel plate ionization chamber, open to the air, made of aluminium 6 mm thick, and surrounded by a 6 mm lead shield (figure 3). The shield filters out much of the low energy radiation, and the aluminium inner liner prevents photo-electrons originating in the lead from causing ionization in the chamber so
that practically all ionization is produced by Compton electrons from the aluminium. Thus the chamber discriminates against low-energy $\gamma$ radiation. Absorption in the radium and in the walls of its container are minimized. Nevertheless, absorption corrections usually have to be applied and these are determined from experimental absorption curves (figure 4).
The electrometer circuit has been improved recently and is probably worth describing (figure 5). A Lindemann electrometer is used in a null balancing circuit. As is usual the change in voltage across the chamber, produced by the ionization current, is balanced by a voltage from a potentiometer. This potentiometer is rotated by a synchronous motor. After 1 minute the end of the potentiometer is reached and the chamber is automatically grounded. Only a known fraction of this steadily decreasing voltage is applied to the chamber through a dekavider, a commercial, very accurate 10,000 step potentiometer. It is only necessary to set the appropriate voltage fraction on the dekavider in order to obtain balance. Ratio determination between two sources can be made to an accuracy of ± 0.2% within a very short time.

The National Research Council participated in several international intercomparisons of radium standards but it is not necessary to go into this since a committee of the International Commission on Radiological Units has recently dealt with this in great detail. It is sufficient to say that the Hönigschmid standards were found to have a relative accuracy of ± 0.2% and this is also the accuracy Hönigschmid claimed for his weighings.

This laboratory still measures about 100 medical radium preparations per year. Each of these preparations is radiographed and figure 6 shows as an example a medical platinum-iridium needle containing two cells filled with radium salt. On receipt of these radiographs an immediate indication is given on the location of the radium within the needle.

In the medical field the radium needles are slowly being replaced by similar needles containing Co\textsuperscript{60}. These are also measured on the same radium measurement apparatus after it has been calibrated for Co\textsuperscript{60}. This was done in the following manner:

With a good, air-equivalent ionization chamber the γ-ray emission of a Co\textsuperscript{60} source in milliroentgens per hour at one meter was determined. Then, this Co\textsuperscript{60} source was compared with a radium standard on our radium measurement apparatus. In this way, and this applies only to our particular ionization chamber, we found that one milligramm radium-equivalent of Co\textsuperscript{60} delivers 0.72 milliroentgens per hour at one meter. We are therefore able to calibrate any other Co\textsuperscript{60} source in these units. Sometimes the effective content of the source in millicuries is wanted; for conversion we use the generally accepted
value for the specific γ-ray emission for Co$^{60}$ of 1.29 milliroentgens per hour per millicurie at one meter.

Many other γ emitters have been standardized by dosimetry directly, i.e. by measuring the exposure dose rate at a certain distance. The value for the specific γ-ray emission which is a particular constant for each γ emitter is used to convert into millicuries. This constant can be calculated when the disintegration scheme of the nuclide concerned is known. Of course, self-absorption in the source should be low and it should be possible to consider the source to be a point source.

Often the activity of the sources is too low to use air-equivalent ionization chambers for the exposure dose measurement. However, a Geiger-Müller counter is, when its walls are made of brass, a quite suitable and much more sensitive device. Since its sensitivity is nearly proportional to the γ energy the counting rate will be a measure of exposure dose. This applies within an energy region of from 0.3 to 3 MeV and the counter method can therefore be used for most γ emitters.

To obtain absolute readings such counters are calibrated with radium, using the recently accepted value for the specific γ-ray emission of 0.825 milliroentgens per hour at 1 meter for 1 milligram of radium enclosed in 0.5 mm platinum. Further checks on the energy response were made with other γ standards.

This method is, of course, rather crude and accuracies of ±10% or better may be achieved. The technique however is very simple and can be used in many cases when the sample cannot be conveniently prepared for
4πβ counting. This is often the case when the sample cannot be destroyed, such as encapsulated radiography sources, or when the specific activity of the radioactive material is too low.

The 4πβ counting method however is probably one of the most important tools for standardization. We have built 4πβ methane flow-counters, identical to those the people in Chalk River have developed and which Dr. Campion has described in the first part of this paper. Figure 7 shows the version of a 4πβ-γ coincidence counter built at the National Research Council. This uses a commercial lead shield and two 2" NaI crystals, one below and one above the 4πβ counter. The upper photomultiplier and crystal assembly can be moved up and down to allow opening of the 4πβ counter for introduction of the source.

Since it seemed that a sufficient amount of gold on the VYNS film is very important to obtain a good counting plateau we have used a simple densitometer to measure the thickness of the gold deposit on the film. Such a densitometer can be obtained commercially; it consists mainly of a light source and a photocell. Figure 8 shows that there is a nearly linear relationship between light transmission density, expressed as log 1/transmission, and the density of the gold. After the transmission measurement the amount of gold was determined chemically by titration with dithizone.

In this connection it is rather interesting to mention that Dr. Baerg from this laboratory recently made some resistivity measurements on the
gold plated films. For ease of handling the films were placed on a polystyrene block. Figure 9 shows the result: the resistivity increases markedly for gold thicknesses below 20 micrograms/cm². For a thickness of about 8 micrograms/cm², the resistivity is $10^9$ times that of bulk gold. It should be mentioned that the resistivities shown here were measured after the films were heated under an infra-red lamp for about 30 minutes at 20 cm distance. In all cases the resistance changed by factors of 2 to 20 upon heating, depending mainly on the age of the film. Figure 10 gives two representative examples, an increase of the resistivity for rather thin films and a decrease for the

![Figure 8](image)

*Figure 8*
Optical density versus thickness of gold deposit on VYNS films

![Figure 9](image)

*Figure 9*
Specific resistance of gold deposit versus thickness
heavier films. It seems difficult to explain this phenomenon: for the low densities isolated crystalites may be formed from amorphous gold with a resulting increase in resistance; however, for higher densities a continuous crystalline structure may build up from amorphous gold, resulting in a resistance decrease.

A crude estimate of the minimum gold thickness required to obtain sufficient conductivity to make a proportional counter work properly was also made. For usual working conditions this would be 6 micrograms/cm², in good agreement with the minimum thickness reported in the literature to obtain counting plateaux.

A fair amount of neutron standardization work is also carried out at the National Research Council. A later paper* will deal with this subject in some detail.

* Paper III. 2., page 393.
I. 2. L'ÉTALONNAGE DES RADIOÉLÉMENTS EN FRANCE

B. GRINBERG
LABORATOIRE DE MESURES DES RADIOÉLÉMENTS DU CENTRE D'ETUDES NUCLEAIRES, SACLAY

Abstract

Calibration of Radionuclides in France

The French laboratories for the measurement of the radioactivity: allocation of work, activities, equipment and staff; future prospects.

Calibration of radionuclides, standard solutions and solid sources: methods of preparation, comparison with corresponding laboratories in other countries, supply statistics.

Étalonnage des radioéléments en France

Les laboratoires de mesures radioactives français: répartition des tâches, activité, équipement et personnels respectifs; perspectives d’avenir.

Étalonnage des radioéléments, solutions et sources solides étalons: techniques de préparation, intercomparaison avec les laboratoires correspondants de différents pays, statistiques des livraisons.

Калибровка радиоэлементов во Франции

Французские лаборатории радиоактивных измерений: распределение заданий, деятельность, оборудование и соответствующий персонал; перспективы на будущее.

Калибровка служащих эталонами радиоэлементов, растворов и твердых источников: методы приготовления, сравнение между соответствующими лабораториями различных стран, статистика поставок.

Normalización de los radioelementos en Francia

El autor describe los laboratorios franceses de mediciones radioactivas: distribución de tareas, actividad, equipo y personal respectivos; perspectivas para el futuro.

También describe el calibrado de radioelementos, soluciones y fuentes patrón sólidas: técnicas de preparación, comparación con los patrones preparados en los laboratorios correspondientes de diferentes países; la memoria termina con datos estadísticos sobre las entregas de radioelementos.

Il existe actuellement en France trois laboratoires officiellement chargés de l'étalonnage des radioéléments et s’occupant de mesures d’activité. Il s'agit de laboratoires dont le rôle essentiel est de délivrer des étalons aux utilisateurs
intéressés ou de faire pour eux des étalonnages de radioisotopes. Il existe naturellement, au sein de divers organismes, d'autres laboratoires s'occupant de métrologie radioactive, mais pour le compte uniquement de ces organismes. Il ne sera parlé dans cet exposé que des trois premiers laboratoires.

I. Service des Mesures de l'Institut du Radium

Ce service fut officiellement créé en 1912, mais il faut dire que dès le début de la découverte du Radium, Pierre et Marie Curie, André Debierne et plus tard Maurice Curie sont venus en aide à l'industrie naissante par leurs conseils, et par la mesure des produits. Cette ouverture officielle a été précédée de la préparation des étalons par Mme Curie et M. Debierne. L'étalon de 16,74 mg de Ra fut accepté en mars 1912 comme étalon international par la Commission des étalons auprès du Congrès international. Cinq étalons de ce laboratoire ont été ensuite comparés à l'étalon international pour assurer l'exactitude des attestations du Service des Mesures. Ces étalons faisaient en août 1911 respectivement: 1,386 mg, 5,41 mg, 19,42 mg, 62,88 mg et 136,0 mg.

La Commission des étalons choisit en outre un étalon auxiliaire de 31,17 mg de RaCl₂ préparé par Höngschmidt pour l'Institut du Radium de Vienne.

Il fut également décidé que les différents étalons secondaires construits pour les besoins des différents pays seraient simplement comparés, par l'intermédiaire du rayonnement γ extérieur, aux deux étalons pesés précités. Les conditions de ces comparaisons furent précisées: utilisation du grand condensateur Marie Curie-Debierne, avec filtration de 1 cm de plomb. A la suite de mesures effectuées successivement à Paris par rapport à l'étalon international, puis à Vienne par rapport à l'étalon auxiliaire, les certificats étaient signés par les deux directeurs des laboratoires précités et par le Président de la Commission des Etalons de Radium.

En 1934, la Commission adopta un nouvel étalon pesé préparé par Höngschmidt (étalon N° 5430) dont la garde et l'utilisation fut confiée au Bureau International des Poids et Mesures.

En 1948 la Commission des Etalons et Unités Radioactives se transforme en Commission Mixte des Etalons et des Unités de Radioactivité. L'étalon international fut placé sous le contrôle de cette nouvelle commission qui en confia la conservation au Bureau International des Poids et Mesures, les mesures de comparaison des étalons secondaires à l'étalon primaire restant assurées par le Laboratoire Curie.

Mais des travaux mirent en évidence une incertitude quant à la définition de la masse de Radium-élément contenu dans l'étalon 5430. La Commission Mixte chargea Mme Joliot-Curie, qui dirigeait le Laboratoire de l'Institut du Radium, de procéder à de nouvelles séries de comparaisons d'étalons Höngschmidt. Un travail important fut fait en collaboration avec le Professeur Kipfer, mais les résultats n'ont pu être publiés.

La Commission mixte a été dissoute en 1956 à Genève et de ce fait il n'est plus possible actuellement de délivrer des certificats d'étalons secondaires.

L'Institut du Radium possède donc actuellement l'étalon Höngschmidt 5430 et l'ancien étalon international "Marie Curie".
En ce qui concerne les moyens de mesure, l’Institut du Radium dispose :
— des méthodes nouvelles de comparaison par le rayonnement γ extérieur, dues à Mme Joliot-Curie et à M. Kipfer ;
— d’un appareillage de fabrication de sources de Radon, indispensables dans ce genre de mesures ;
— d’une méthode de micro-calorimétrie adiabatique à compensation automatique mise au point dans cet Institut.

Il serait désirable que la situation incertaine dans le domaine de l’étalonnage des sources de Radium soit clarifiée. L’Institut du Radium, actuellement dirigé par M. le Professeur Teillac, souhaiterait que la garde de l’étalon international et la comparaison de cet étalon avec les étalons nationaux, rôle qui lui était dévolu jusqu’en 1957, revienne au Bureau International des Poids et Mesures. L’Institut du Radium, en raison des moyens dont il dispose, de l’expérience acquise dans ce domaine pourrait naturellement apporter sa collaboration aux tâches que le B. I. P. M. se fixerait.

Outre l’étalonnage des sources de Radium, dans les conditions qui viennent d’être sommairement décrites, le Service de Mesure de l’Institut du Radium effectuait, pour le compte de divers utilisateurs, des mesures de radio-éléments naturels sous des formes variées. Ces mesures étaient sanctionnées par un certificat. Depuis le début de 1959 ce type d’activité a été confié au Service de Radioactivité du Laboratoire National d’Essais, service récemment créé, par suite d’une convention passée entre ce Laboratoire et l’Institut du Radium. Le matériel correspondant à ce type de mesures ainsi qu’un étalon secondaire de Radium ont été remis par le Laboratoire Curie au Laboratoire National d’Essais.

II. Laboratoire de Mesures des Radioéléments du Centre d’Etudes Nucléaires de Saclay

L’activité de ce laboratoire, de création relativement récente (1952), est essentiellement centrée sur la mesure et l’étalonnage des radioéléments artificiels. L’étalonnage des radioéléments naturels et en particulier du Radium n’est pas de son ressort, en principe au moins. Mais il arrive cependant, occasionnellement, que des mesures de radioéléments naturels lui soient confiées.

Le laboratoire de mesures, créé à l’origine exclusivement pour satisfaire les besoins internes du Commissariat à l’Énergie Atomique a progressivement été conduit à satisfaire les besoins d’utilisateurs extérieurs. Disposant seul jusqu’ici des moyens nécessaires, ce laboratoire exerce pour la France, en ce qui concerne la standardisation des radioéléments artificiels, une activité comparable à celle du National Physical Laboratory en Grande Bretagne, du National Bureau of Standards aux États-Unis, de l’Institut Mendeleïev en Union Soviétique, du Physikalische Bundesanstalt en République Fédérale Allemande, etc. . . .

Son rôle dans ce domaine est de livrer des solutions ou des sources étalonnées et certifiées aux laboratoires qui en font la demande et de procéder à des intercomparaisons internationales.

En tant qu’organisme du C. E. A., le laboratoire effectue des mesures
variées pour le compte de divers services, en particulier les mesures de solutions ou de sources solides délivrées régulièrement à divers utilisateurs.

1. Solutions et sources étalonnées

Le laboratoire délivre périodiquement des solutions étalonnées des radio-éléments courants (P32, Co60, I131 . . .). Il prépare en outre, sur demande, des solutions étalonnées de la plupart des radioéléments qui figurent sur le catalogue du C. E. A. Les activités livrées sont soit de l'ordre du µc/cc soit du mc/cc. La précision, qui dépend du radioélément, varie de 1 à 3%. Le laboratoire délivre également des sources étalons de dimensions et de présentation standardisées:
— sur film mince montée sur un anneau
— sur support rigide en plexiglass
— sur support rigide et recouverte d'un couvercle protecteur.

Lorsque cela est possible, en particulier pour les radioéléments à vie longue, une partie des solutions étalonnées est conservée dans un dispositif de stockage. On dispose ainsi d'une réserve de solutions étalons qui permet, d'une part de satisfaire les demandeurs sans délai, d'autre part de vérifier périodiquement la stabilité des solutions.

Il s'est avéré nécessaire de contrôler la pureté radiochimique des solutions et de déterminer leur teneur, ou tout au moins la limite supérieure de celle-ci en impuretés radioactives. Les moyens utilisés à cette fin sont l'absorption $\beta$ et la spectrométrie $\gamma$, complétés prochainement par deux dispositifs de mesure de la période, à l'aide de chambres d'ionisation différentielles.

<table>
<thead>
<tr>
<th></th>
<th>1957</th>
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<th>1959 (9 premiers mois)</th>
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<tr>
<td>Solutions délivrées</td>
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<td>118</td>
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</tr>
<tr>
<td>Sources</td>
<td>1</td>
<td>111</td>
<td>177</td>
</tr>
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</table>

Répartition des livraisons entre les radioéléments en 1959

<table>
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<tr>
<th></th>
<th>Solutions</th>
<th>Sources</th>
</tr>
</thead>
<tbody>
<tr>
<td>Na$^{24}$</td>
<td>7</td>
<td>3</td>
</tr>
<tr>
<td>P$^{32}$</td>
<td>23</td>
<td>26</td>
</tr>
<tr>
<td>S$^{35}$</td>
<td>11</td>
<td>18</td>
</tr>
<tr>
<td>Co$^{60}$</td>
<td>17</td>
<td>27</td>
</tr>
<tr>
<td>Se$^{80}$ + Y$^{90}$</td>
<td>27</td>
<td>54</td>
</tr>
<tr>
<td>Cs$^{137}$</td>
<td>18</td>
<td>39</td>
</tr>
<tr>
<td>I$^{131}$</td>
<td>17</td>
<td>1</td>
</tr>
<tr>
<td>Au$^{198}$</td>
<td>8</td>
<td>8</td>
</tr>
</tbody>
</table>

2. Techniques Utilisées

Je ne donnerai dans ce qui suit qu'un aperçu rapide des principales méthodes lorsqu'elles sont classiques et un peu plus détaillé pour celles développées récemment.
A. Emetteurs β

1° Compteur 4π G. M. — C'est l'appareil fondamental pour l'étalonnage des solutions. Le modèle mis au point au Laboratoire de Mesures (figure 1 a et 1 b), associé à un réservoir de 30 litres, comporte un “sas” permettant d'introduire ou d'extraire les sources. Le compteur peut ainsi fonctionner plus d'un mois, à raison de 10 à 20 mesures par jour, sans renouvellement du gaz.

**Quelques caractéristiques:**

gaz: Argon (8 cm) + Propane (1.5 cm)
supports de sources: films minces d'acétate de cellulose non métallisé à graphite incorporé, de 5 à 10 μg/cm²; pour les émetteurs peu énergiques, de mylar.

Sources: de l'ordre de 2 à 3 mg, pesées à la microbalance.
Lorsque la solution contient des quantités appréciables d'entraîneur, on utilise un mouillant: Tween 20, dilution 1/25.
2° Méthode de l’angle solide défini.
3° Coincidences β-γ.
4° Méthodes d’étalonnage secondaire:

a) Chambre d’ionisation associée à un électromètre Wulff bifilaire (figure 2). L’utilisation de récipients, dont le fond est constitué par un film mince, disposé au dessus de la chambre permet de mesurer aussi bien les émetteurs mous que les émetteurs énergiques.

Seuils de mesure: 1 μc/cc pour P³²
20 μc/cc pour S³⁵.
Les activités mesurées peuvent atteindre 40 mc/cc pour P³².

b) Dispositif à faible mouvement propre et grande géométrie.

Ce dispositif est décrit dans la communication présentée à ce colloque par Y. Le Gallic, B. Grinberg et M. Thénard*. Il permet de déceler des activités à partir de 10⁻⁵ μc/cc pour le C¹⁴ et de 10⁻⁵ μc/cc pour le P³², donc nettement inférieures, même dans le cas le plus défavorable (Sr⁹⁰) aux activités permises. La sensibilité, les possibilités de la méthode ne sont affectées ni par la nature du solvant, ni par la présence de substances inactives en solution.

* Paper II. 2. 5., page 245.
c) Chambre d'ionisation $\beta$-$\gamma$.
Cette chambre, qui peut servir à la mesure de solutions d'activités supérieures à 0.1 mc/ml, est utilisée principalement à la mesure des applicateurs utilisés en médecine (films plastiques marqués). Le seuil de mesure en ce cas est de 60 $\mu$C/cm$^2$.

B. Emetteurs $\alpha$
Dans la plupart des cas l'émission est mesurée en 2$\pi$ par des chambres d'ionisation.

![Figure 3]

a) Chambre à impulsion à air. D'un emploi très commode, son utilisation est toutefois limitée à des activités inférieures à $5 \times 10^{-8}$ $\mu$C en raison, en particulier, du temps de résolution relativement élevé.

b) Chambre à grille à remplissage d'Argon + 2,5% Azote à la pression de 2,5 kg/cm$^2$. Cet appareil convient aux activités comprises entre $10^{-6}$ et 0,2 $\mu$C. Associée à un sélecteur à 100 canaux cette chambre est également utilisée pour la spectrométrie $\alpha$ (figure 3).

c) Chambre d'ionisation à courant. Elle est utilisée pour la mesure de dépôts de très faible épaisseur et de forte activité, par exemple
de Plutonium. Cette chambre, placée dans une enceinte étanche peut facilement être commandée à distance. La gamme d’activités mesurables va de 0.1 μc à 500 μc.

C. Émetteurs γ

Pour les sources solides utilisées en gammographie, pour les solutions déjà flaconnées et prêtes pour la livraison, il est nécessaire de prévoir un contrôle non destructif. Le laboratoire dispose à cette fin d’un certain nombre d’appareils.

a) Chambre d’ionisation à puits (figure 4 a et 4 b).

Cette chambre remplie d’Argon sous pression est utilisable pour une très large gamme d’activités: les valeurs extrêmes sont dans le rapport 1/10⁶.

Pour une source ponctuelle la zone d’égale sensibilité — à ± 1% — est sensiblement un cylindre de 4 cm de diamètre et 11 cm de hauteur. Ceci est dû à ce que la distance interélectrode n’est pas constante mais réduite dans la région médiane par l’adjonction aux électrodes de manchons cylindriques. De
ce fait l'étalonnage établi pour une source ponctuelle reste valable pour des sources de dimensions finies que l'on peut donc mesurer dans des conditions satisfaisantes. Le seuil de mesure est d'environ 0,1 μc pour le Co\(^{60}\) (sensibilité pour Co\(^{60}\): \(6 \times 10^{-13}\) A/μc). Par commutation des résistances d'entrée de l'amplificateur associé on peut mesurer la gamme de courants d'ionisation correspondants. Cet appareil a depuis peu été commercialisé.

b) **Chambres air-équivalentes**

Elles ne nécessitent pas d'étalonnage préalable pour chaque radioélément; il suffit de connaître le "facteur K spécifique". Deux types sont utilisés:

1\(^{o}\) chambre en graphite de 50 cm\(^3\) associée à un dispositif électrométrique constitué par un électromètre Compton et un condensateur étalon. La mesure est faite par la méthode de Townsend. L'ensemble du dispositif est réalisé de façon telle que les γ diffusés ne puissent intervenir dans la mesure. Le seuil mesurable est de l'ordre du mc (figure 5).

2\(^{o}\) Chambre à puits en graphite. Elle réalise la synthèse des deux précédentes: sa réponse en roentgens est indépendante de l'énergie des γ et son
efficacité est élevée. Ici encore la forme des électrodes a été calculée de façon à avoir à l'intérieur du puits un volume de sensibilité constante aussi grand que possible. Pour des dimensions extérieures de 9 cm de diamètre et 16 cm de hauteur la zone d'égale sensibilité est un cylindre de 6,5 cm de hauteur et 1,5 cm de diamètre (figure 6).

Figure 5

Des manchons de graphite amovibles, d'épaisseurs variées, permettent de tracer la courbe du courant d'ionisation en fonction de l'épaisseur et d'extrapoler à zéro.

La mesure se fait par comparaison à un étalon de Radium en utilisant le rapport des facteurs $K$ des deux sources. La sensibilité pour le Radium est de $0,75 \times 10^{-11} \text{ A/mc}$.

Les dispositifs qui viennent d'être énumérés et sommairement décrits sont les principaux de ceux dont le Laboratoire de Mesures dispose présentement. On met en outre au point actuellement la mesure des gaz radioactifs par comptage interne et la mesure de sources $\alpha$ intenses par microcalorimétrie.
III. Laboratoire de Radioactivité du Laboratoire National d'Essais

Ce laboratoire national est appelé à relayer le laboratoire du Centre de Saclay pour une part de ses activités. De création relativement récente (début 1959) il ne pourra aborder l'étalonnage des radioéléments artificiels que lorsqu'aura été mis en place l'équipement nécessaire. Il jouera alors — pour la France — le même rôle que les divers laboratoires nationaux étrangers spécialisés dans ce domaine. Présentement son activité se limite aux mesures dont il a été parlé dans le chapitre I de cet exposé.
I. 3. STANDARDIZATION OF RADIONUCLIDES IN THE ELECTROTECHNICAL LABORATORY — TOKYO

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Abstract

Since 1953, the Electrotechnical Laboratory has undertaken absolute measurements of the activities of some radionuclides by means of the $4\pi\beta$ counting method and the coincidence method.

The details of the counting methods used in our laboratory will be described.

1. $4\pi\beta$ Counting Method — a bell type of $4\pi\beta$ gas proportional counting apparatus was designed, and its counting characteristics were investigated, especially the dependence upon the counting gases. In measuring the activity, about 50 mg of the sample was taken on a thin film and then dried in the silica gel desiccator. One drop of a wetting solution was added on the dried sample so as to reduce the self-absorption, and the sample was dried again. The thin film was prepared through the procedure that synthetic resin (Kanevinyl-R1108 in cyclohexanon) spread over the water surface was taken up on the metal ring and then covered with very thin gold by means of the vacuum evaporating method. Sr$^{85}$-Y$^{90}$, Co$^{60}$ and Cs$^{137}$ were measured by the above procedure. The accuracies of the measurements were of the order of ±2%.  

2. $2\pi\alpha$ Counting Method — The samples, RaD + E + F, U$^{238}$ and Po$^{210}$ were measured by means of the same apparatus as in the $4\pi\beta$ counting method. The accuracies of the measurements were ±2, ±1 and ±1% respectively.

3. Coincidence Counting Method — In the coincidence method, both $\beta$-$\gamma$ and $\gamma$-$\gamma$ counting methods were employed. A scintillation counter [NaI (Tl), $1'' \times 1''$] was used as $\gamma$-ray detector, and a GM counter as $\beta$-ray detector. Recently, in order to examine the inherent difference between the $4\pi\beta$ counting and the coincidence counting methods, instead of the GM counter, we devised a special type of the $4\pi\beta$ proportional counter, which can also be used as $4\pi\beta$ proportional counting apparatus independently, apart from the coincidence circuits. The results obtained from the $\beta$-$\gamma$ coincidence method were, in general, somewhat different (3—4%) from those of $4\pi\beta$ counting method. This may be due to the self-absorption. On the other hand, the result for Co$^{60}$ obtained from $\gamma$-$\gamma$ coincidence method was in agreement with that of $\beta$-$\gamma$ coincidence counting method.

Étalonnage des radionuclides au Laboratoire d'électrotechnique de Tokyo

Depuis 1953, le Laboratoire d'électrotechnique fait des mesures absolues de certains émetteurs par les méthodes de comptage $4\pi\beta$ et de comptage par coincidences. Les auteurs donnent les détails ci-après sur les méthodes de comptage appliquées dans ce laboratoire:
1. Comptage $4 \pi \beta$. — Le laboratoire a mis au point un compteur-cloche $4 \pi \beta$ proportionnel; il a étudié ses caractéristiques, en fonction notamment du gaz de comptage employé. Une très petite quantité, 50 mg environ, de l'échantillon est déposée sur un film support mince, et séchée ensuite dans un exsiccateur contenant du gel de silice. Une goutte de solution de détergent synthétique est ajoutée à l'échantillon, après séchage, afin de réduire l'autoabsorption, puis celui-ci est séché à nouveau.

Le film support mince est préparé de la façon suivante: une goutte de résine synthétique (Kanevinyl-R 1108 dissoute dans le cyclohexane), déposée à la surface de l'eau, donne une pellicule fine qui est recueillie sur l'anneau de métal puis recouverte d'une très mince couche d'or par la méthode d'évaporation sous vide. Les nuclides Sr$^{90}$-Y$^{90}$, Co$^{60}$ et Cs$^{137}$ ont été mesurés par la méthode ci-dessus. La précision des mesures à été de l'ordre de $\pm 2\%$.

2. Comptage $2 \alpha$. — Les échantillons de RaD + RaE + RaF, U$^{238}$ et Po$^{210}$ ont été mesurés à l'aide de l'appareil décrit sous 1. La précision des mesures a été de $\pm 2$, $\pm 1$ et $\pm 1\%$, respectivement.

3. Comptage par coïncidences. — Le comptage $\beta$-$\gamma$ et le comptage $\gamma$-$\gamma$ ont été tout deux employés. Un compteur à scintillations [cristal de NaI activé au thallium, d'un pouce de diamètre et d'un pouce de longueur] a été utilisé comme détecteur $\gamma$ et un compteur G.M. comme détecteur $\beta$. Récemment, pour établir la différence entre les méthodes de comptage $4 \pi \beta$ et de comptage par coïncidences, les auteurs ont remplacé le compteur G.M. par un compteur proportionnel $4 \pi \beta$ spécial qui peut être utilisé aussi, indépendamment du circuit à coïncidences, comme compteur $4 \pi \beta$. Les résultats du comptage par coïncidences $\beta$-$\gamma$ ont été en général quelque peu différents (3 à 4\%) de ceux du comptage $4 \pi \beta$. Cette différence peut provenir de l'autoabsorption. En revanche, pour le Co$^{60}$, le résultat du comptage par coïncidences $\gamma$-$\gamma$ a confirmé celui du comptage par coïncidences $\beta$-$\gamma$.

Стандартизация радиоизотопов в электротехнической лаборатории в Токио

После 1953 года в электротехнической лаборатории с помощью вычислительного метода $4 \pi \beta$-счёта и метода совпадений было произведено абсолютное измерение некоторых изотопов.

Предварительный метод вычислений в нашей лаборатории будут описаны в следующем порядке:

1) Метод $4 \pi \beta$-счета. Был сконструирован звонковый газовый пропорциональный счетчик $4 \pi \beta$-излучений. Были исследованы его вычислительные характеристики, в особенности надежность таких применяемых газов. При измерении активности было взято очень маленькое количество (50 мг) образца на тонкой пленке. Оно было высушено в силикагельном вакуум-котле. Затем образец был вычеркан снова.

Тонкая пленка была получена в результате того, что синтетическая смола, покрывавшая водную поверхность, была перенесена на металлическое кольцо и затем покрыта очень тонким слоем золота с помощью выпаривания в вакууме. Изотопы Sr$^{90}$ — Y$^{90}$, Co$^{60}$ и Cs$^{137}$ были измерены вышеупомянутым способом. Точность измерений составляла $\pm 2\%$.

2) Метод $2 \alpha$-счета. Образцы RaD + E + F, U$^{238}$ и Po$^{210}$ были измерены с помощью тех же аппаратов, которые применялись при методе $4 \pi \beta$-счета. Точность измерения составляла $\pm 2$, $\pm 1$ и $\pm 1\%$ соответственно.
3) Вычисление методом, основанном на принципе совпадений. При применении этого метода были использованы методы β-γ и γ-γ счета. Сцинтилляционный счетчик [NaI (TI), 1" × 1"] был использован в качестве детектора γ-лучей, а счетчик Гейгера-Мюллера — как детектор β-лучей. Недавно для того чтобы исследовать существующее различие между вычислением методом 4 π β-счета и методом, основанном на принципе совпадений, мы вместо счетчика Гейгера-Мюллера сконструировали специальный тип пропорционального 4 π β-счетчика, который может, кроме того, быть использован в качестве пропорционального 4 π β-счетчика независимо от схем совпадений. Результаты, полученные методом, основанным на принципе совпадений, для β-γ излучений в общем несколько отличались (на 3—4%) от результатов метода вычисления 4 π β-счета. Это может произойти благодаря самопоглощению. С другой стороны, результат для Co60, полученный за счет метода, основанного на принципе совпадений для γ-γ излучений, согласовался с вычислением с помощью подобного же метода для β-γ излучений.

Normalización de radionúclidos en el Laboratorio de Electrotécnica de Tokio

Desde 1953, se vienen efectuando en el Laboratorio de Electrotécnica de Tokio mediciones absolutas de las radiactividades de algunos núclidos utilizando el método de recuento β 4 π y el de coincidencias.

A continuación se describen los métodos de recuento seguidos en el Laboratorio.

1) Recuento β 4 π. — Se ha construido un contador proporcional de gas β 4 π, de tipo campana y se han estudiado sus características de recuento, en particular los efectos de los gases utilizados en el recuento. Para medir la radiactividad se deposita una cantidad muy pequeña de la muestra (unos 50 mg) sobre una película delgada, secándola a continuación en un desecador que contiene gel de sílico. Sobre la muestra seca se añade una gota de una solución de detergente sintético para reducir la autoabsorción y la muestra se vuelve a secar. La película se obtiene por el siguiente procedimiento: sobre la superficie del agua se vierte una resina sintética (Kanevinyl-R1108 disuelta en ciclohexanona) que se monta sobre el anillo metálico y se recubre con una delgada capa de oro por el procedimiento de evaporación al vacío. Utilizando este método se midió la actividad de los núclidos Sr90-Y90, Co60 y Cs137. La precisión de las mediciones fue del orden de ± 2%.

2) Recuento α2 π. — Se midió la radiactividad de muestras de RaD + RaE + RaF, U238 y Po210 utilizando el mismo aparato que en el método de recuento β 4 π. La precisión de las mediciones fue de ± 2, ± 1 y ± 1%, respectivamente.

3) Recuento por coincidencias. — Para aplicar este método, se emplearon aparatos de recuento β-γ y γ-γ. Como detector de rayos γ se utilizó un contador de centeleo [cristal de NaI (TI) de 1 pulgada de diámetro y 1 pulgada de largo] y como detector de rayos β un contador Geiger-Müller. Recientemente, para estudiar las diferencias existentes entre el método de recuento β 4 π y el de coincidencias, en lugar de utilizar un contador Geiger-Müller, los autores recurrieron a un modelo especial de contador proporcional β 4 π, que puede ser también empleado independientemente, es decir, sin los circuitos de coincidencia. Los resultados obtenidos con el método de coinci-
dencias $\beta-\gamma$ difieren ligeramente ($3-4\%$) de los alcanzados con el método $\beta 4 \pi$. La discrepancia puede deberse a la autoabsorción. En cambio, los resultados del método de coincidencias $\gamma-\gamma$ para el Co$^{60}$ concuerdan con los obtenidos al aplicar el método de coincidencias $\beta-\gamma$.

1. The Electrotechnical Laboratory is responsible for the standardization of radionuclides and the maintenance of radioactive standards in Japan. Therefore, since 1953, we have investigated the absolute measurement of radionuclides by $4 \pi \beta$ counting, by $2 \pi \alpha$ counting and coincidence methods. We have calibrated standard sources and supplied several kinds of radioactive standard solutions in ampoules.

The standard solutions already prepared are indicated in Table I.

**TABLE I**

<table>
<thead>
<tr>
<th>Radiation</th>
<th>Nuclide</th>
<th>Normal activity</th>
<th>Volume</th>
<th>Carrier solution</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\gamma (\beta)$</td>
<td>Co$^{60}$</td>
<td>$10^6$ d. p. s.</td>
<td>5 ml</td>
<td>CoCl$_2$ 1 g/l in 0.1 N HCl</td>
</tr>
<tr>
<td>$\gamma (\beta)$</td>
<td>Co$^{60}$</td>
<td>$10^5$ d. p. s.</td>
<td>5 ml</td>
<td>CoCl$_2$ 1 g/l in 0.1 N HCl</td>
</tr>
<tr>
<td>$\beta (\gamma)$</td>
<td>Co$^{60}$</td>
<td>$10^4$ d. p. s.</td>
<td>ca 5 ml</td>
<td>CoCl$_2$ 0.5 g/C in 0.1 N HCl</td>
</tr>
<tr>
<td>$\beta$</td>
<td>Sr$^{90} + \gamma$</td>
<td>$10^4$ d. p. s.</td>
<td>ca 5 ml</td>
<td>SrCl$_2$ 0.016 g/l</td>
</tr>
<tr>
<td>$\gamma (\beta)$</td>
<td>Cs$^{137}$</td>
<td>$10^6$ d. p. s.</td>
<td>5 ml</td>
<td>YCl$_3$ 0.016 g/l in 0.1 N HCl</td>
</tr>
<tr>
<td>$\gamma (\beta)$</td>
<td>Cs$^{137}$</td>
<td>$10^5$ d. p. s.</td>
<td>5 ml</td>
<td>CsCl 1 g/l in 0.1 N HCl</td>
</tr>
<tr>
<td>$\beta (\gamma)$</td>
<td>Cs$^{138}$</td>
<td>$10^4$ d. p. s.</td>
<td>ca 5 ml</td>
<td>CsCl 0.05 g/l in 0.1 N HCl</td>
</tr>
</tbody>
</table>

Up till now we have had no opportunity of comparing the accuracy of our measurements with those of other national institutions. We have measured the secondary radioactive standards available commercially from other countries, for example the NBS in USA, employing our instruments for standardization.

In this report, our methods, instruments and results of standardization will be described.

2. $4 \pi \beta$ counting:

(a) Arrangement.

The most direct method to determine the disintegration rate of $\beta$ emitters is to count all $\beta$ particles emitted per disintegration. The $4 \pi$ counter aims at this purpose. Figure 1 indicates the construction of our $4 \pi$ counter. It consists of two $2 \pi$ counters, and a sample is introduced onto the surface of the boundary by mounting on a thin film. The anodes are 0.04 mm $\Theta$ tungsten wire. The minimum distance between either anode and the sample is about 12 mm. The film used for mounting the sample is part of the cathode.

Each $2 \pi$ counter is respectively connected to its preamplifier, the output signals of which are added in parallel, amplified by a main amplifier, discriminated, and finally counted by a scaler. The $4 \pi$ counter is operated in the proportional region. The main amplifier used is a modified Atomic Inst. Co model 204B linear amplifier. The dynamic range is over five thousand, the
gain about ten thousand and the sensitivity at the input is 2 mV. The scaler is a Tracer Lab. Scaler 1000 (resolving time, 5 μsec). The stabilized high electrode voltage (4000—5000 V) is supplied from the variable high voltage source utilizing high frequency oscillation.

(b) Procedure.

The measuring sample is mounted on a thin polyvinyl acetate film supported on a thin metal ring, the internal diameter of which is 16 mm. One surface of the film is coated with evaporated gold. The total thickness of the film is about 25 μg/cm². The polyvinyl film is prepared as follows: one part (by volume) of polyvinyl acetate (Kanevinyl R 1108) is dissolved in 10 parts of cyclohexanon, and a small quantity of this solution is placed on the edge of a glass slide, from which it is spread on the surface of water, solidified, scooped up by a metal ring and finally dried in air. Such a film is very uniform in thickness. In preparing the measuring sample, about 50 mg of radioactive solution is dropped on the polyvinyl film by a micropipette and dried. The method of drying varies according to the nuclide.

As counting gases, "Q", "PR", methane and propane gases were examined. Methane and propane gases are used for fuel and are not pure. These gases, except "Q" gas, gave perfect horizontal plateau, proving no counting loss. In fact, methane gas is suitable according to cost and the operating voltage.

3. 2πα counting.

2πα counting of RaF (in radioactive equilibrium with RaD, RaE on a silver disk faced with a palladium layer was generally used as a β-ray standard) was tried for estimating the disintegration rate of RaD. The advantage of the above method lies in the simplicity of the corrections due to the
self-absorption and the back-scattering of α-rays. To perform $2\pi$ α counting, one half of the $4\pi$ counter is used in the α counting region, namely, input sensitivity of the amplifier — 28 mV; applied voltage — 3000—3100 V. The necessary procedure to find out the disintegration rate of RaD, is to estimate the geometry, self-absorption, and the ratio of the disintegration rates of RaD and RaF. Thus, three aged RaD + E + F NBS standards have been measured for one year and a half. Though the maximum difference between our experimental results and the certificate values of NBS was 5.2%, the accuracy of our measurement seems to be ± 2%. Furthermore, NBS standard samples, U$_2$Os, Po$^{210}$ were measured by $2\pi$ α counting, and these results are indicated in Table II.

### TABLE II
Results of $2\pi$ Counting

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Po$^{210}$</th>
<th>RaD + E + F</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample No</td>
<td>B-10</td>
<td>D-3</td>
</tr>
<tr>
<td>NBS Certified Value</td>
<td>176 ± 2%</td>
<td>484 ± 2%</td>
</tr>
<tr>
<td>ETL Certified Value</td>
<td>179 ± 1%</td>
<td>494 ± 1%</td>
</tr>
</tbody>
</table>

These values are corrected for decay to reference date.

4. Coincidence counting.

(a) Introduction.

Coincidence counting methods are very useful for the absolute measurement of radioactivities. In our laboratory, the study of this method was initiated at the same time as $4\pi$ counting. Up to the present, we have made absolute measurements of Co$^{60}$ and Na$^{24}$. The coincidence counting methods employed are of three kinds: β-γ, γ-γ, and $4\pi$ β-γ coincidence counting. The last is a sort of β-γ coincidence counting. According to this method, not only the coincidence counts are increased, but also $4\pi$ β counting may be simultaneously carried out with an identical sample. Therefore, it is a method for finding out the difference of the results by both methods.

(b) Instruments.

The γ-ray detector consists of a 1" X 1" NaI (TI) crystal and a 6291 photomultiplier tube. The β detector is a G-M counter (argon-alcohol, mica end window, mica thickness: 1.7 mg/cm$^2$) or a $4\pi$ β counter, which is almost the same as the $4\pi$ β counter above mentioned. The amplifier of the scintillation counter has a 0.1 μsec. rise time, and delay-line clipping time of 0.5 μsec. The pulses from each channel to the coincidence mixer are shaped into square waves by the univibrator delay-line circuit in order to fix the resolving time. It was found that there was no counting loss under the following conditions:
for the $\beta$ channel a pulse width of 1 $\mu$sec; and
for the $\gamma$ channel a pulse width of 1 $\mu$sec when the GM tube is used as
a $\beta$ counter, 3 $\mu$sec when the 4 $\pi$ counter is used as a $\beta$ counter.
The samples are prepared by mounting on thin plastic plates, the diameter
of the sample itself being about 4 mm $\phi$, since, it is difficult to reduce further
the size of the sample without sacrificial the weighing precision. For 4 $\pi$ $\beta$-$\gamma$
coincidence counting, sample preparation is the same as for 4 $\pi$ $\beta$ counting.
In $\beta$-$\gamma$ coincidence counting the GM tube and the scintillator are separated
by 10—14 cm and the sample is placed midway between them. The back-
ground coincidence counting rate is 0.75 cpm. The $\gamma$-ray detection efficiency
of the GM counter is found by placing an aluminium absorber having a
suitable thickness in front of the window of the GM counter. The position
of the absorber is determined by reducing the $\gamma$-ray count to a minimum.
For $\gamma$-$\gamma$ coincidence counting, two scintillation counters of the above type
are used. In the case of Co$^{60}$, the angle between both scintillation counters
is 130°, considering the angular correlation.

5. Results of 4 $\pi$ and coincidence counting
(a) Results of 4 $\pi$ counting.
(i) Sr$^{90}$—Y$^{90}$.
The sample was prepared from a NBS standard sample ($\neq$ 4910,
10610 $\pm$ 2% dps/ml on 4.15.1953) by drying in ammonia vapour. The
corrected value for decay by assuming the half-life of Sr$^{90}$ as 25 years, is
10850 dps/ml on 4.15.1953 which is 2.3% higher than the certified value
of NBS.
(ii) Tl$^{204}$.
The sample was prepared from a Nuclear Chicago Corporation standard
sample (RS-204, $0.191 \times 10^5 \pm 3\%$ dps/ml on 6.7.1957) by drying in
ammonia vapour. The measured value was corrected to 12046 $\pm 3\%$ dps/ml
on 9.4.1959 by estimating the absorption loss as 1%, and counting loss due
to the electron capture as 2.4%.
(iii) Cs$^{137}$.
A part of Cs$^{137}$ stock solution (1 mg/ml CsCl, 0.1N HCl) was diluted
twenty times with 0.1N HCl, then dried with a small volume of detergent
solution. This treatment gives a very reproducible and high counting rate. By
estimating the corrections due to the $\gamma$-ray efficiency of Ba$^{137m}$ as 0.4%, the
absorption loss as 0.5% and internal conversion as 9.1%, the activity of the
stock solution was found to be $2.002 \times 10^5 \pm 2\%$ dps/ml on 2.1.1958.
We tried with a scintillation counter the mutual $\gamma$-ray comparison of
our Cs$^{137}$ standard sample, the NBS standard sample and the English (Radio-
chemical Centre) standard sample, all of which were in glass ampoules. It
becomes clear that our evaluation is 0.8% higher than the English and 9.2% higher than the NBS.
(b) The result of coincidence counting of Co$^{60}$.
The stock solution (1 mg/ml CoCl$_3$, 0.1N HCl) was found to be $2.87 \times 10^6$
$\pm 3\%$ dps/ml on 9.4.1959 by $\beta$-$\gamma$, $\gamma$-$\gamma$ coincidence counting. From 4 $\pi$ $\beta$-$\gamma$
coincidence counting, $2.86 \times 10^6 \pm 3\%$ dps/ml on 9.4.1959 was obtained
but $4\pi \beta$ counts only gave a value about 3\% lower than the above. It is thought that this difference is probably due to the self-absorption of low energy $\beta$ particles.

By the intercomparison of our Co$^{60}$ standard sample with NBS standard sample ($0.685 \times 10^6 \pm 2\%$ dps on 10.1.1953) our stock solution was calibrated as $0.285 \times 10^6$ dps/ml on the basis of standard, assuming the half life of Co$^{60}$ as 5.3 years.
I. 4. ROUTINE STANDARDIZATION OF RADIONUCLIDES IN THE UK

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Abstract

Standards of radionuclides, whose activity has been measured either directly by a primary method, or by comparison with a primary standard, are available from the NPL and the AERE. Measurements of standards are carried on also at other laboratories such as RCC and AWRE. Some estimate of the number of staff employed and of standards issued will be given. An advisory committee, of those directly interested in this work at the institutions named and one or two others, has promoted the interchange of information and measurements.

For routine issues, NPL uses mainly $4\pi\beta$ counting in its various forms, linked with coincidence counting, and reference ionization chamber methods. To deal with a wider variety of nuclides, AERE uses gas and scintillation counting for special purposes. The apparatus employed will be described and the internal consistency, and agreement with the results of other laboratories, which has been attained over the past few years, will be discussed, for a number of the more important nuclides.

At the original suggestion of the Committee, a combined $\beta-\gamma$ reference ionization chamber has been designed and is now commercially available; many of these chambers have been calibrated at NPL, and their performance and consistency will be discussed.

Méthodes courantes d'étalonnage des radionuclides au Royaume-Uni

Des radionuclides étalonnés, dont l'activité a été mesurée soit directement, soit par référence à un autre étalon, sont en vente au National Physical Laboratory (NPL) et à l'Atomic Energy Research Establishment (AERE). L'étalonnage de radionuclides se fait également dans d'autres laboratoires comme le Radiochemical Centre (RCC) et l'Atomic Weapons Research Establishment (AWRE). L'auteur donne une estimation des effectifs employés et du nombre des étalons préparés. Un comité consultatif composé de personnes qui, dans les établissements susmentionnés ainsi que dans un ou deux autres, s'intéressent directement à ces travaux, a facilité l'échange de renseignements et de mesures.

Pour la préparation d'étalons d'usage courant, le NPL utilise surtout les diverses formes de compteurs $4\pi\beta$, associés à des dispositifs à coïncidences, ainsi que des chambres d'ionisation. Pour l'étalonnage d'autres nucléides, l'AERE utilise des compteurs à gaz et à scintillations spéciaux. L'auteur décrit les appareils employés et indique, pour un certain nombre des nucléides les plus importants, dans quelle mesure les résultats obtenus depuis quelques années concordent entre eux et avec ceux obtenus par d'autres laboratoires.

En réponse à une proposition du comité, on a mis au point une chambre d'ionisation combinée $\beta-\gamma$, actuellement en vente dans le commerce; un grand nombre de chambres de ce type ont été étalonnées au NPL; l'auteur étudie leur comportement et leur fidélité.
Стандартизация радиоизотопов обычными методами в Великобритании

Стандарты радиоизотопов, активность которых была измерена либо непосредственно первичным способом, либо сравнением с первичным стандартом, могут быть получены из Национальной физической лаборатории Англии (НФЛ) или Научно-исследовательского центра по атомной энергии (Харвилл, Англия). Измерения стандартов производятся также и в других лабораториях таких, как РСС и Научно-исследовательский центр по разработке атомного оружия (Англия). Будут даны некоторые сведения о численности занятого персонала и выпущенных стандартах. Консультативный комитет, состоящий из тех, кто непосредственно заинтересован в этой работе в перечисленных учреждениях и в одном или двух других, способствовал обмену информацией и результатами измерений.

Для обычных измерений НФЛ использует, как правило, $4\pi \beta$-счет в его разнообразных формах, связанных с методом вычисления, основанным на принципе совпадений, и методом исследований с помощью контрольной ионизационной камеры. Для специальных целей при работе с более разнообразными видами изотопов Научно-исследовательский центр по атомной энергии использует газовые и сцинтиляционные методы счета для специальных целей. Будет описана аппаратура, применяемая при этом, и для некоторых более важных изотопов будут обсуждены внутреннее соответствие и согласованность по отношению к результатам других лабораторий, полученных в течение последних нескольких лет.

По первоначальному предложению Комитета сконструирована и в настоящее время доступна для приобретения комбинированная контрольная ионизационная камера для $\beta$ и $\gamma$ излучения; многие из этих камер калибровались в Национальной физической лаборатории, и их работа и надежность подлежат рассмотрению в последующее время.

Normalización de radionúclidos en el Reino Unido

El National Physical Laboratory y el Atomic Energy Research Establishment pueden proporcionar patrones de radionúclidos cuyas actividades se han medido directamente por un método primario, o por comparación con un patrón primario. También se mide la actividad de patrones en otros laboratorios, como el Radiochemical Centre y el Atomic Weapons Research Establishment. Es autor da algunos datos sobre el personal empleado y los patrones preparados. Un comité consultivo, integrado por las personas que se interesan directamente por estos trabajos en los centros mencionados y por algunas otras personas, ha fomentado el intercambio de información y de resultados de las mediciones.

De ordinario, el National Physical Laboratory utiliza sobre todo métodos de recuento $\beta 4\pi$ en sus diversas formas, complementados con procedimientos de recuento mediante dispositivos de coincidencias y de cámaras de ionización. Para la normalización de otros radionúclidos, el Atomic Energy Research Establishment utiliza en casos especiales contadores de gas y de centelleo. El autor describe los aparatos empleados y, para algunos de los núcleidos más importantes, indica la medida en que los resultados obtenidos en el curso de los últimos años concuerdan entre sí y con los datos a conocer por otros laboratorios.
A propuesta del Comité, se ha diseñado una cámara de ionización de referencia β-γ combinada; esta cámara ya se ha puesto a la venta. Muchas de estas cámaras han sido calibradas en el National Physical Laboratory; el autor estudia su rendimiento y características de recuento.

It is important, in talking about "standards", to know the meaning we attach to this term. The careful distinctions drawn by McNISH (1), for example between "prototype", "derived", etc. standards, are difficult to apply to the radioactive case. Our basic unit, the curie, is now defined as a number of disintegrations per second, so that even the international and national radium standards of the Höngschildt series have, in a sense, only a comparative rather than an absolute value. Any laboratory which is prepared to put enough skill and effort into such processes as $4\pi$ counting, so as to improve on the present techniques, can set up, if it wishes, its own radioactive standards, more precise than those of the national laboratories.

In fact, however, this does not happen. Users of radionuclides are anxious to avoid the trouble, and are happy to acquire standards from specialist laboratories, either standard samples of known disintegration rate, or standard apparatus so designed that they can measure their own samples with sufficient accuracy on an absolute basis. The degree of accuracy required itself varies widely. The biological or medical worker may not be able to detect a 5% or 10% variation in the radiation effect produced with which he is concerned. The nuclear physicist can probably, as in all fundamental work, make use of all possible accuracy in the acquisition of new knowledge. The reactor engineer can point out that, in a contract involving the measurement of radioactive performance, an error of 1% might correspond with a differential cost of a million pounds.

In fact, while comparative ionization, calorimetric or counting measurements may often be made to an accuracy of 1 in $10^3$, the absolute disintegration rate of a given radioactive sample is rarely measurable to an accuracy better than 1 in $10^2$. This accuracy is sufficient for most present-day users; but it remains as a challenge to the physicist, who continuously seeks to improve it.

In the United Kingdom, there are three laboratories from which workers with radionuclides can obtain "standard" material — using the term in its widest sense, to mean material to which a measurement process, with some pretensions to accuracy, has been applied. They are:

(a) The National Physical Laboratory

The NPL is formally responsible for radioactive standards in the UK, and it deals with international comparisons. It distributes standardized solutions of certain radionuclides (at present, Na$^{24}$, P$^{32}$, I$^{131}$, Au$^{198}$, Sr$^{90}$+$Y^{90}$, Co$^{60}$) at stated intervals (e.g., once a year); these are in sealed ampoules of standard size, in concentrations varying from 1 μc/gm to 1 mc/gm. It is the custodian of the national (Hönigschildt) radium standard, and in special circumstances will make comparisons, by γ-ray ionization, of radium sources with this or related standards. It has sponsored the manufacture and sale by a commercial firm of the chamber type 1383A, which is a combined β-γ
ionization chamber having stable and repeatable characteristics; and it will calibrate such chambers on request. It has recently installed equipment for the comparison of neutron sources, and is prepared to calibrate such sources against the national standard source which is just being set up.

The work at NPL has for many years had the benefit of a special advisory committee, formed from those personally most active in radionuclide measurements in the United Kingdom. The committee has circulated standards for measurement among its members, and the NPL has not placed any radionuclide on its list of standards issues until the Committee has agreed that the accuracy and consistency of measurement obtainable with this particular nuclide are satisfactory.

The limits of accuracy quoted on the NPL certificates are conservative (usually ± 2% for Co⁶⁰, ± 5% for I¹³¹, and ± 3% for the other nuclides); the consistency, within the Laboratory, of a given standardization is much better than this (a S. D. for example of 0.2 to 0.5% is typical); but experience of international and national comparisons over a period of years indicates that the true confidence limits must not be set too narrowly.

In a typical year (1958) NPL issued 142 radionuclide standards, 19% of which were for international comparison; 65% were requested by research laboratories and 16% by hospital workers.

(b) The Atomic Energy Research Establishment

The AERE will make measurements on some 60 other radionuclides, on request, which are not available through NPL. The accuracies quoted (ranging from ± 2% to ± 5%) assume the validity of published decay schemes, and in general the limits are wider for the soft β emitters.

Many measurements are made at AERE for use within their own organization. In a typical year, some 300 sources are measured in absolute terms, and a further 175 sources are prepared from calibrated solutions. Perhaps half of these sources are for use within the Establishment; and about half the remainder are distributed overseas.

(c) The Radiochemical Centre

is the main channel in the UK for the distribution of radioactive material of all kinds, including specially labelled compounds, and industrial radioactive sources. The activities of their materials are mainly quoted as "nominal" or "minimum" values. The Centre however supplies a large variety of "Radioactive Reference Sources" which are intended mainly for checking counting equipment. α, β, γ and neutron solid sources are available; also dilute solutions (e. g. of Cs¹³⁷, I¹³¹, P⁸⁰, Sr⁹⁰) for calibrating liquid counters. Accuracies of intercomparison with the Centre's own standards (themselves often maintained by comparison with NPL standards) are quoted over a wide range, from 2% to 15%.

The demand for these reference sources is considerable; already in 1959 the Centre has distributed approximately 100 liquid and 400 solid β sources, 100 α sources, and 200 γ sources.

I will now refer in a little more detail to the methods used in these standard measurements. All the three laboratories concerned use 4π β counting techniques, and coincidence counting techniques, for a majority
of their measurements; and their equipment is basically very similar. The details which I quote here are those of NPL practice. In a typical NPL $4\pi\beta$ proportional counting set-up, the counter is of the twin-hemisphere design with a loop electrode in each hemisphere. 90% argon 10% methane flows through the counter at atmospheric pressure, and a plateau of 250 volts is usually obtained. Counting at 6 points along the plateau, each of over 10$^5$ counts, gives a statistical deviation of about 0.1%. Corrections are applied for paralysis-time (5 μsec, 0.5%), background (0.2%; a bias level referred to the input of 50 μV is used), source-mount absorption (no correction is at present applied, using gold plated VYNS mounts, 20 μg/cm$^2$). The amplifiers are of the Chase-Higginbotham non-overloading type.

For coincidence counting, the source is placed in a cylindrical $4\pi\beta$ counter, which has a thin-walled base; close to this is the NaI-thallium counting crystal. The γ count rate is about $1/20$th of the β count rate, and the background up to 8% of the γ count rate. Sufficient counts are taken to give a statistical error of 0.3% in the coincidence channel. Corrections are applied for paralysis time (for the β channel 5 μsec, up to 1.5%; for the γ and coincidence channels 15 μsec, up to 0.15%, background, and accidental coincidences (perhaps 6%; determined experimentally). No correction is applied for the γ sensitivity of the β counter, in view of its high β detection efficiency.

Gas counting is not in use at the NPL (except for the special work of the radiocarbon dating unit). At AERE it is used in calibration of tritium and C$^{14}$.

Liquid scintillation counting is used at RCC for tritium and C$^{14}$ calibrations and occasionally at AERE for tritium.

Calorimetric methods, in spite of the excellent example set by MANN(4) at NBS, have not yet been used in the United Kingdom for routine standardization.

Ionization chambers are used particularly at the NPL as a check on the continuity of direct counting methods. A simple graphite-walled cylindrical chamber, with Lindemann electrometer and Townsend induction balance, gives great stability and reliability. Readings are reduced to NTP values; the screen round the lead from the chamber to the balance is exhausted of air. A γ-ray source is measured here by direct comparison with a standard radium source. Other simple but robust ionization chambers, one with approximately $4\pi$ geometry for γ-ray measurement e.g. from a standard ampoule, and another for $2\pi$ measurement from solid sources on nickel dishes, are of great value in rapidly checking a process of dilution, a series of similar sources, etc.

I will speak in rather more detail about the combined β-γ ionization chamber, which in the view of the NPL Advisory Committee on Radioactive Standards, to which I have already referred, should provide for many workers a simple, cheap and useful means of standardization. Figure 1 shows the construction of the chamber. It was designed so that, without great expense (the current price of this chamber is some £ 80), it could be manufactured to give constant characteristics; in fact, for γ sources the chambers as supplied by the manufacturers have sensitivities within about ± 1%, the corresponding
limits for a selection of $\beta$ sources being about $\pm 3\%$. Results obtained with one chamber are thus applicable to others of the same type, provided that a uniform procedure for source preparation and measurement has been adopted.

If a closer calibration is needed than the $\pm 1\%$ or $\pm 3\%$ above mentioned, these individual chambers can be measured at the NPL, on request, by comparison with the reference chamber of this type which it maintains.

Construction of the chamber is clear from the diagram. It is normally operated with a polarizing voltage of 300 V, giving saturation within $0.3\%$ at $5 \times 10^{-9}$ amp. (equivalent to a $\text{Co}^{60}$ source of about 100 mc). Currents may be measured by any suitable DC amplifier (e.g. the 1388 A amplifier manufactured by Messrs. Avo Ltd): the background current is of the order $10^{-14}$ amp. The chamber is not sealed; corrections are normally made to NTP on an air-density basis, modified by an empirical factor which has been determined for a number of radionuclides. If the $\beta$ window (duralumin, $0.68 \text{ mg/cm}^2$) is broken, it can be replaced by one supplied by the manufacturers without appreciable alteration in calibration.

Data are available also on the effects of screening, the effects of variation of size and position of sources, and on the measured currents per mc given by a variety of nuclides. (cf. Table I).
<table>
<thead>
<tr>
<th>Radioactive isotope</th>
<th>Ionization current</th>
<th>Radioactive isotope</th>
<th>Ionization current</th>
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<tr>
<td></td>
<td>γ-ray chamber</td>
<td>β-ray chamber</td>
<td></td>
</tr>
<tr>
<td>Na$^{24}$</td>
<td>(A $\times$ 10$^{-12}$ per mc$^1$)</td>
<td>(A $\times$ 10$^{-12}$ per mc/ml)</td>
<td>Br$^{82}$</td>
</tr>
<tr>
<td>Ps$^{82}$</td>
<td>(b) 75.5</td>
<td>(c) 268</td>
<td>(a) 63</td>
</tr>
<tr>
<td>K$^{42}$</td>
<td>(b) 6.0</td>
<td>(c) 433</td>
<td>(b) 62</td>
</tr>
<tr>
<td>Fe$^{56}$</td>
<td>(a) 27.1</td>
<td>(d) 53.2</td>
<td>(c) 103</td>
</tr>
<tr>
<td>Co$^{60}$</td>
<td>(a) 53.2</td>
<td>(b) 53.2</td>
<td>(a) 10.6</td>
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<tr>
<td></td>
<td>(d) 53.2</td>
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<td>(c) 335</td>
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<td>(c) 73</td>
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<td>(c) 137</td>
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<td></td>
<td>Ra$^{226}$ + decay products</td>
<td>(A $\times$ 10$^{-12}$ per mg)</td>
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<td></td>
<td></td>
<td>(b) 35.6</td>
<td>(b) 35.6</td>
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<tr>
<td></td>
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<td>(c) 34.5</td>
<td>(c) 34.5</td>
</tr>
</tbody>
</table>

* Bremsstrahlung.

(a) 1 g of solution in a 2 ml ampoule.
(b) 4 g of solution in a 5 ml ampoule (each supported in the source cavity in the appropriate perspex holder).
(c) 1 ml of solution in a polythene dish.
(d) Co$^{60}$ in an AERE Birmabright capsule, in the holder used for 2 ml ampoules; flat end of capsule on base of holder; long axis of capsule vertical.
(e) Radium source (active length 3 mm, in platinum tube 6 mm long having walls 0.5 mm thick); flat end of tube on base of holder; long axis of tube vertical.

Though neutron source standardization on a routine basis has so far not been available in the UK, I should like to say a little about the equipment and methods which are just now being set up at NPL for this purpose. We had two initial tasks (a) to acquire a satisfactory technique for the comparison of similar neutron sources (b) to satisfy ourselves as to the absolute accuracy of measurement of a particular neutron source, to be set aside as a British National standard.

There are two types of method so far suggested for the absolute calibration of neutron sources (a) those in which the fast neutrons from the source are moderated in a tank or block of hydrogen-containing material, and then, when at thermal velocities, captured and measured or (b) those in which some "associated" particle can be measured; e.g. in the usual radium-beryllium source reaction, $^{9}$Be$^8 + \gamma = n + 2$He$^4$, and it should be possible to measure the neutrons produced from the number of helium atoms also set free.

In this work, NPL has the advantage of close collaboration with AERE and with Mr. Martin at Durham University. Six Ra-Be sources have been intercompared among themselves (by the Mn$^{56}$ activity induced in a tank of manganese solution, also by Bf$3$ counters embedded in polythene); some had previously been compared with other sources (e.g. those used by Oxford, Harwell and NBS). An internal consistency of order 0.1% has been obtained.
in the NPL intercomparisons. An absolute measurement has also been completed, by the manganese method; and it is hoped that soon, at Durham University, the helium content of three of these sources will be measured, giving an absolute determination by a quite separate method.

I have now surveyed very briefly the routine standardization measurements which are made in the UK, though I have said nothing about the standardization of radium itself, almost entirely carried out at the RCC by normal γ-ray ionization measurement; about weak radium standards, on which in the past NPL has done some work; nor about precise α standards, for which at present there seems to be little demand in the UK.

What of the future of radionuclide standardization? Developments could be in (a) the provision of a more numerous list of available standardized nuclides (b) a significant increase in absolute accuracy.

The present list of standards obtainable from the NPL, at specified times in the year, is — Na$_{24}$, P$_{32}$, I$_{131}$, Au$_{198}$, Sr$_{90}$, Co$_{60}$. For all these, there have been intercomparisons between several laboratories, and agreement that a satisfactory state of experience and technique has been reached, before standard samples have been issued under the NPL certificate. We are trying to extend this list to include also Na$_{22}$, K$_{42}$, Cs$_{137}$ and Tl$_{204}$. The work on Na$_{22}$, at least, is sufficient for its issue to be agreed shortly.

More generally, there can be very few nuclides of technical interest for which standardisation, of a kind, has not yet been provided. There are however significant classes of nuclides (e. g. electron-capture nuclides, and those with very low energy β emission) where the straightforward methods available at present can give considerable errors. For how many such nuclides is it really going to be worth while to develop absolute standardization methods, to an accuracy of 1 or 2%? — still more, to undertake specialized research to extend this accuracy into the 1 in $10^3$ region? We have to remember that for many purposes, comparative measurements only are needed; and simple methods can give accurate comparisons for the majority of nuclides. As experience, for example, with the NPL β-γ ionization chamber builds up in different laboratories, we hope to have a really comprehensive list of calibration factors, covering a wide range of β and γ energies, available for the user.

As to (b), there are so many links in the chain of absolute calibration that there will always be possibilities for future refinement of accuracy. On the counting process itself, there is in a sense no limit of accuracy — except perhaps with nuclides of very short half-life. The statistical errors of the measurement are easily determinable. But we are often uncertain about (a) the efficiency of the equipment (absorption in the mount, self-absorption in the source, dead-time background, volumetric efficiency in gas-counting) (b) what is really being measured (purity of sample, dilution and absorption errors, uncertain chemistry, dubious decay-schemes, or cross-sections).

To conclude this paper, therefore, I shall trace in a little more detail the history of our calibration at NPL of a single radionuclide, I$_{131}$. I have chosen this one in particular because it is one of the most important from the user’s point of view, and its standardization has a longer history than any. Also it is one of the most difficult nuclides with which to obtain consistent results. Earlier measurements between different laboratories have on occasions shown discrepancies of several per cent. The β spectrum is not highly energetic, so
that there may be a considerable foil correction to apply. Also it is possible to lose iodine, if sources are prepared simply by the evaporation of the usual KI solution. It has been NPL practice in preparing the source to precipitate the drop immediately by adding silver nitrate to it; without this technique, losses as big as 10% might easily occur.

A provisional I$^{131}$ standard was set up, at the original request of the Medical Research Council, as long ago as 1948. It was based on the measurement of γ ionization in air in known conditions, assuming a knowledge of the disintegration scheme, the γ-ray absorption coefficients, and the energy to produce an ion pair. A little time after this, the NPL assumed the maintenance of this standard. Meanwhile, more exact knowledge about the basic assumptions had been accumulating and the technique of counting methods had improved. In 1953 it was decided that the best value, from the work of several laboratories, taking into account counting methods as well as ionization methods, was given by taking an I$^{131}$ conversion factor of 2.25 r/hr at 1 cm for 1 mc. (The 1948 value was 2.54, i.e. a jump of 13%.) This figure was published as a British standard, and standard samples have been distributed since then on the basis of ionization measurements, using the graphite chamber which has already been described.

Whenever I$^{131}$ standards have been issued by NPL, however, simultaneous measurements have been made (a) on the 4πγ ionization reference chamber and (b) by 4πβ counting, using, both aluminium and VYNS source mounts.

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**Figure 2**

**IODINE $^{131}$ – N.P.L. STANDARDS**

- X Ratio Graphite / 4πγ Ionization
- □□ Ratio 4πβ counting (Al foil) / 4πγ Ionization
- ○○ Ratio 4πβ counting (VYNS) / 4πγ Ionization

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...
Figure 2 shows the ratio of the two ionization measurements over this period of six years. The standard deviation of this ratio is ± 0.22%, indicating a very satisfactory consistency of the equipment. Figure 2 shows in addition the ratios of the β counting measurements to the standard ionization measurement. You see here the fluctuations in value, which are rather larger than we should like to see; and I think these results justify our policy of maintaining the standard of this particular nuclide by ionization rather than by counting methods.

Finally, figure 3 shows the data of the previous figure recalculated on the following assumptions: — that K (Radium) is 8.25 r/hr/mg at 1 cm (instead of 8.3), that W is 33.7 eV (instead of 32.5) — leading to a value of K (I^{131}) of 2.209 instead of 2.25; that the absorption correction for aluminium foil mounts should be unchanged at 2.2%, but that for the VYNS foils, a correction of 0.8% should be applied to cover foil plus source absorption (there is some separate evidence that this is about the right figure).

You will see that the effect of these changes is to narrow the gap between the three groups of data, though of course leaving unchanged the capricious fluctuations in the 4πβ counting data. If we were to adopt them finally, it would mean a change in the British I^{131} standard of 0.4% — hardly I think justified in present circumstances.

To give a proper assessment of the absolute accuracy with which we know such a standard as I^{131}, I should have to refer to the results of the many
international comparisons which have taken place in the past years; and so should be going outside the title of this paper. But I have said enough, I think, to justify the conservative limits, of plus or minus several per cent, which we still feel it desirable to attach to our calibrations of radionuclides on an absolute basis.

Acknowledgments

My thanks are due to my colleagues in the Radiology Section, NPL, for helpful discussions and for the data presented in this paper. Much of the work referred to forms part of the research programme of the NPL, and it is described by permission of the Director of the Laboratory.

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I. 5. ABSOLUTE MEASUREMENT OF RADIOACTIVE MATERIALS AT THE PHYSIKALISCH-TECHNISCHE BUNDESANSTALT

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Abstract

A short report is given about the routine methods of standardization of radionuclides. The measurements are done by $4\pi\beta$ proportional counters, $4\pi\beta$-$\gamma$ coincidence counting and for secondary standardization of $\gamma$ emitters by $4\pi\gamma$ ionization chamber. The equipments used and the procedures of source preparation are described.

Mesure absolue des substances radioactives à l'Institut fédéral de physique appliquée

L'auteur présente un rapport succinct sur les méthodes courantes de normalisation des radionuclides. Les mesures sont effectuées au moyen de compteurs proportionnels $4\pi\beta$, par voie de comptage par coïncidences $4\pi\beta$-$\gamma$ et, en ce qui concerne la normalisation secondaire des émetteurs de rayons $\gamma$, au moyen d'une chambre d'ionisation $4\pi\gamma$. Le mémoire contient une description du matériel utilisé et des procédés employés pour la préparation des sources.

Абсолютное измерениеadioактивных веществ в физико-техническом федеральном институте

Приводится краткий доклад по обычным методам стандартизации радиоизотопов. Измерения осуществлялись $\beta$-пропорциональными счетчиками на $4\pi$, счетчиками $\beta$-$\gamma$ совпадений и $\gamma$-ионизирующей камерой на $4\mu$ для вторичной стандартизации $\gamma$-излучателей. Приводится описание используемого оборудования и процедура подготовки источника.

Determinación absoluta de la actividad de sustancias radiactivas en el Instituto Federal de Física Aplicada

En la memoria se describen brevemente los métodos corrientes de calibración de radionúclidos. Las mediciones a que se refiere se han efectuado con contadores proporcionales $\beta$ $4\pi$, contadores de coincidencias $\beta$-$\gamma$ $4\pi$; para la calibración secundaria de emisores $\gamma$ se ha utilizado una cámara de ionización $\gamma$ $4\pi$. Se describen los aparatos empleados y los procedimientos de preparación de las fuentes.

In this report a short survey is given on the standardization of artificially produced radioactive nuclides, carried out at the laboratory for radioactivity of the Physikalisch-Technische Bundesanstalt (PTB) Braunschweig (Federal Republic of Germany) during the last three years. These investigations were
accomplished especially in view of preparation and distribution of standard solutions of such radioisotopes being used frequently for medical and biological applications. The delivery of the short-lived radionuclides Na$^{24}$, P$^{32}$, I$^{131}$ and Au$^{198}$ is scheduled to begin within the next year. Standard solutions of the long-lived nuclides Co$^{60}$ and Sr$^{90}$— in equilibrium with Y$^{90}$— shall be available from stock. Standard solutions of Ra$^{226}$ with a content of $4.10^{-9}$ g Ra-element for radon analysis have been issued by the PTB during the last 4 decades.

Although the basic methods are nearly the same as those employed in most of the other laboratories, i.e. $4\pi \beta$ counting and $\beta$-γ coincidence counting, it may appear worth while to give a brief report here.

$4\pi \beta$ counting

Initially $4\pi \beta$ measurements were done with $4\pi$ counters operated in the Geiger-Müller region. The counters were filled to a total pressure of 10 cm Hg with argon and an admixture of 10% ethanol. Using a quenching circuit similar to that described by PUTMAN$^1$ good plateaus were achieved with slopes less than 1% per 100 V over a region of 200 V. The results obtained were in a good agreement with those by $4\pi \beta$ proportional counting in our and other laboratories. No systematic errors were found. The usual standard deviation was $\pm 0.4\%$.

Measurements with Geiger-Müller counters have the advantage that the electronic layout is straightforward. On the other hand this method is time consuming because of pulse-rate limitations due to dead-time losses and because the counter has to be evacuated and refilled after each change of sample. This is therefore why $4\pi \beta$ measurements are currently done with flow counters operated in the proportional region.

The construction of one of the counter types currently in use is shown in figure 1. Each half consists of a modified "pill-box" type. In order to avoid regions of reduced electrical field strength, pieces of aluminium are inserted, forming cathodes of semi-cylindrical shape. In comparison with the formerly used pill-box counters, better plateaus have been achieved with the present design. A 0.05 mm stainless steel wire, extending between teflon insulators, forms the anode. The two halves of the counter are separated by a slider containing two holes for source insertion. A gastight seal of the slider is effected by two "0" rings, mounted in each half. This construction avoids the necessity to open the counter directly to the atmosphere when changing the sources. The sources are deposited on thin gold-coated plastic films, which are stretched over rings of aluminium, 40 mm o. d. and 14 mm i. d. Such a source mount is shown in the foreground of figure 1.

Methane of technical purity is used as a flow gas at atmospheric pressure. With both, the Fairstein and the Chase-Higinbotham non-overloading amplifier, good plateaus were obtained between 3200 and 3800 V. As no slope within the statistical error of $\pm 0.1\%$ has been measured, it may be concluded that nearly all emitted $\beta$ particles are detected.

For each standardization a set of 5 to 10 sources is prepared usually with activities ranging from 0.5 to $3.10^5$ dpm. The method of source preparation will be considered later. Each source is measured separately in two different $4\pi$ counters as a check for proper operation of the counters. No systematic deviation beyond normal statistical error has been found yet. Counting is
effected by preset time technique with time intervals of 10 minutes and more
to reduce the statistical fluctuation as well as the error caused by the in-
accuracy of the timer, which is of the order of ± 0.5 sec. As the timer is
driven by a synchronous motor, any deviation in line-frequency results in a
faulty pulse-rate measurement. The accuracy and stability of line-frequency
has been tested during the measurements in comparison to the standard fre-
quency of the PTB and no deviation beyond 0.1% has been found. Therefore
the error in registered counts per unit time is considered to be less than ± 0.1
to ± 2%. Generally, 4 π counting is independent of the complexity of the
decay scheme of the nuclide involved since cascade gamma rays or electrons
from internal conversion add to the original beta ionization pulse if they

![Figure 1](image)

4 π β proportional counter currently in use at the PTB

occur within the resolving time of the detector and electronic circuitry. Hence
no corrections are necessary. Only in the case that a metastable state exists
with a life time long compared to the resolving time, γ-ray efficiency of the
γ counter and internal conversion coefficients must be known accurately and
accounted for, for instance in Cs¹³⁷ standardization.

Apart from this the following factors have to be considered:
1) the counting loss by dead time,
2) the self-absorption in the source,
3) the absorption in the mounting film,
4) the counting loss by the low-energy cut-off and the radial extension
   of the sources.

Dead time can be measured easily by a double-pulse generator or with
a set of sources of known activities, so that the error introduced by dead-time
corrections may be considered to be less than 0.1 to 0.2%.

Self-absorption in the source, however, is the limiting factor in the use
of the 4 π β proportional counting method. It depends not only on the energy
spectrum of the nuclide in question but also on the granularity of the solid source material as well as on the amount of carrier added. Therefore it can be different from measurement to measurement. Comparisons with results obtained by $4\pi\beta-\gamma$ coincidence measurements gave values of self-absorption for Au$^{198}$ and I$^{131}$ of the order of 1 to 2% and for Co$^{60}$ between 3% for chemically treated sources and up to 8% for untreated sources.

The absorption in the mounting films, which generally are of 40 $\mu$g/cm$^2$ in total thickness, is accounted for only in the case of low-energy beta emitters. Since this correction is small, the uncertainty of results caused by absorption in the mounting film can be 0.2%.

The counting efficiency, i.e. probability that a $\beta$ particle emitted will also be detected, is lowered first by the low-energy cut-off and secondly by a decrease in detection efficiency for particles started in the outer hemispheres of the counter. The low-energy cut-off is found in the order of 200 to 300 eV. On account of the energy spectra of the nuclides considered here, the relative number of electrons with energies below this value is smaller than 0.1%, this deficit can be neglected. The geometric dependence of the counting efficiency was measured by moving a point radiator out of the centre of the detector. Constant efficiency was found within a statistical error of ±0.1% over an area 10 mm in diameter. Since the aliquots normally used yield deposits about 6 mm in diameter, there is no correction necessary in this regard.

Apart from self-absorption in the source the total uncertainty in $4\pi$ counting with counting tubes can therefore be considered to be within the limit from 0.2% for the high-energy beta emitters up to about 0.5% for the low-energy beta emitters.

$\beta-\gamma$ coincidence counting

For $\beta$ emitters which are associated with one or more $\gamma$-rays in coincidence, the method of $\beta-\gamma$ coincidence counting is one of the most powerful for absolute standardization since the result is independent from the efficiency of either detector and therefore independent from self-absorption in the source.

The simple $\beta-\gamma$ coincidence arrangement with detectors of low efficiency for the emitted radiations in each channel however, introduces other corrections and uncertainties into the result. So the decay scheme must be known, especially the internal conversion coefficients, as well as the $\gamma$-ray efficiency of the $\beta$ counter. These difficulties can be avoided if $\beta$ detection is effected by a $4\pi$ counter (2). Because of the high efficiency of the $\beta$ detector most corrections are small.

The $4\pi\beta$ counter used in this coincidence arrangement is of the same type as shown in figure 1. Only the slider is somewhat longer in order to facilitate the replacement of samples across the 10 cm lead shield which surrounds both detectors during the measurements. The $\gamma$ detector consists of a NaI (Tl) crystal, 4 in. in diameter and 2 in. high coupled to an EMI 9530 photomultiplier tube.

A block diagram of the electronic circuitry is shown in figure 2. The two channels are identical. The outputs of the detectors are fed each to an amplifier and single channel pulse-height analyzer of the Fairstein type. In the $\beta$ channel the analyzer is operated in “integral” position. Dead time and
resolving time are defined by two pulse-formers and are variable in steps. Generally equal dead times of 3 μsec are used. The resolving time is determined to 1.7 μsec by double-pulse generator and accidental coincidence monitoring.

Generally, the only corrections in 4π β-γ coincidence measurements are those due to background, accidental coincidences and dead-time losses, expressions for which are derived by CAMPION (2). Since the corrections are small and the quantities involved can be measured exactly, results derived from these measurements may be considered accurate within 0.1 to 0.2%.

Source preparation

Several methods and materials were examined for preparing thin and uniform films with appreciable tensile strength. Good results were obtained with celluloid dissolved in isoamyl acetate and "Mowital" — a polyvinylformal — soluted in acetylene dichlorid. The Mowital films show excellent mechanical properties.

The procedures of foil preparation are the following: 300 mg Mowital are dissolved in 100 ml of acetylene dichlorid. A glass plate is put perpendicularly into this solution for about 5 seconds. After removal, the glass plate is coated with a thin, uniform film. Cuts are made along the edges of the plate with a razor blade and then the film is floated off on distilled water and picked up on an aluminium anuli of 14 mm inner diameter and 40 mm outer diameter. Uniform 10 μg/cm² thick foils can be prepared easily by this method. The foils are then coated with a conducting gold layer of 30 μg/cm² by vacuum distillation. Thereafter a drop of insulin is pipetted on to the centre of the foil and resucked immediately leaving back only a very thin layer some 5 mm in diameter.

The weight of the liquid deposit is determined gravimetrically by means of a micro-balance. After measuring the tareweight of the source mount, a liquid aliquot of 5 to 30 mg is pipetted on to the insulin film. In the very moment of pipetting a stopwatch is started and the weight is measured 1, 2 and 3 minutes thereafter. A plot of dropweight against time gives an exact straight line which can be extrapolated to zero time. By this procedure the weight of the liquid aliquots can be determined with an accuracy of better than ±10 μg, that means ±0.1% for an aliquot of 10 mg.

Initially the samples were dried under an infra-red lamp. In measuring a set of sources in two different 4π counters, an individual standard deviation
of $\pm 1\%$ was found between the different sources whereas the reproducibility between the two counters was of the order of $\pm 0.3\%$. So it must be concluded that this discrepancy was caused by an inaccuracy in the weights. Several tests, however, showed that weighing was correct within $\pm 0.1\%$, but the following observation was made: by vehement drying at the end of evaporation, when only a thin layer of solution is left, the rest of the droplet is evaporated as a visible cloud, perhaps taking off part of the active material. In common practice therefore, the samples are dried in open air at room temperature or by gentle heating under an infra-red lamp. By this technique the standard deviations have been reduced to $0.4\%$ or less.

In the cases of $^{131}\text{I}$ and $^{60}\text{Co}$ the samples are treated chemically for precipitation of the active material in the usual manner.

**Relative standardization**

At the Physikalisch-Technische Bundesanstalt the primary or absolute standardization of the short-lived $\gamma$ emitting nuclides is maintained by means of a $4\pi \gamma$ ionization chamber in conjunction with a set of reference standards of radium, consisting of radium salt, flame-sealed in small glass tubes. Primary standards of the short-lived nuclides calibrated by $4\pi \beta$ or $4\pi \beta-\gamma$ measurements are compared with the reference sources in this chamber under defined geometrical conditions. All further standardization is done then against the reference sources by means of the related Ra-equivalence established by the primary comparison.

A schematic drawing showing the construction details of the $4\pi \gamma$ ionization chamber presently in use is presented in figure 3. The chamber consists of a cylindrical steel vessel, 30 cm high and 25 cm in diameter. The sources are inserted into the central cylinder which is 19 cm deep and 3 cm in diameter. Ampoules with solutions of the active material to be measured are supported on a lucite spacer, while the reference sources are suspended into the central cavity by means of small hooks.
The collecting voltage is applied to the outer wall of the chamber whose outer side is painted with a well-insulating lacquer. The insulated collector is connected with a vibrating-reed electrometer by a shielded cable. Current measurements are made in a rate-of-charge null method.

Figure 4 shows the dependence of the ionization current from the position of a point source which is moved a) in vertical and b) in radial direction through the inner cavity. As curve a) shows the ionization current remains nearly constant up to a vertical displacement of 4 cm. However the depth of the inner cylinder is not optimal, a somewhat deeper cavity would result in a longer plateau. Because of the large influence of radial displacements, the sources must be well centred, which can be done easily within ± 2 mm.

Without an outer lead shield the background current is less than \(4 \times 10^{-14}\) A corresponding to a background equivalent activity of 0.3 µg of radium.

The individual standard deviations are of the order of 0.5% and the reproducibility of measurements over the past six months has been better than 0.5%.

Up to the present, the chamber was calibrated with Na\(^{24}\), Co\(^{60}\), I\(^{131}\) and Au\(^{198}\). Figure 5 shows a plot of the radium equivalent per unit \(\gamma\)-ray activity as a function of the mean \(\gamma\) energy of the nuclide involved. Here **\(\gamma\)-ray**
activity” means the genuine activity multiplied by the mean number of γ quantums emitted per disintegration. By interpolation this plot allows the calibration of any γ-ray emitter with γ energies between about 0.3 and 2 MeV without a primary standardization by an absolute measurement, if the decay scheme is well known.

The Physikalisch-Technische Bundesanstalt is glad having had the opportunity of participating in international intercomparisons since the past three years. It is mainly by these measurements, that we have been able to develop our technique to the present state. We are grateful for the efforts of all those, who have made possible this successful cooperation.

Figure 5
Energy response of the 4π γ ionization chamber.

REFERENCES

Radioactive Metrology Methods in the USSR

The authors review the methods practised in the USSR and in particular at the Mendeleev Institute of Metrology at Leningrad, for the reproduction, by means of standardized apparatus, of the units of measurement used for radioactivity (curie, gramme-equivalent of Ra, roentgen and rad).

As regards the radionuclides, the standard sources regularly produced in the USSR are of two types:

1. Model sources, which are used in standardizing other radioactive substances by comparison and in calibrating radiometric and monitoring instruments; and

2. Control sources, which are used exclusively for checking the functioning of measuring instruments and the reproducibility of the data yielded by them.

Model α sources are prepared by electrolytically depositing suitable selected emitters (natural U, U$^{233}$, Pu$^{239}$ and Am$^{241}$) on platinum. Activities over the range 30—10$^6$ disintegrations per minute can be provided. The control α sources consist of an electrolytic deposit of Pu$^{239}$ in a layer with a specific weight of between 3.10$^{-9}$ and 0.7 mg/cm$^2$, corresponding to activities of 10—10$^8$ disintegrations per minute.

Model β sources are made of radionuclides selected so as to provide a range of maximum energies of 0.018—2.3 MeV.

Control β sources are prepared from either Sr$^{90}$ + Y$^{90}$, Tl$^{204}$ or Pm$^{147}$. These sources cover a wide range of activities of between 20 and 10$^9$ disintegrations per minute.

Model γ sources are made of either Ra, Co$^{60}$ or Cs$^{137}$. The range of activities covered by these standards falls between the orders of magnitude of 10$^{-11}$ and 5 gramme-equivalents of Ra.

The control γ sources are based on the same radionuclides as mentioned above. Their activities cover a very wide range and they vary very considerably in form and size.

Lastly, the model neutron sources are of two types: Ra-α-Be (containing 1—1000 mg Ra) and Pu-α-Be (containing 0.01—15 g Pu). The Ra-α-Be sources consist of a compressed mixture of RaBr$_2$ and powdered Be with a Ra/Be ratio of 1 : 6 by weight. The Pu-α-Be sources consist of an alloy (PuBe$_{13}$) of these two metals.
En ce qui concerne les radionuclides, les sources étalons couramment produites en URSS sont de deux types:

1° Sources modèles servant à l'étalonnage par comparaison d'autres préparations radioactives, ainsi qu'au calibrage d'appareils radiométriques et de moniteurs de rayonnement;

2° Sources de contrôle servant exclusivement à la vérification du fonctionnement et au contrôle de la reproductibilité des indications fournies par les appareils de mesure.

Les sources α modèles sont préparées par dépôt électrolytique sur platine d'émetteurs convenablement choisis (U naturel, U_{233}, Pu_{239}, Am_{241}). Peuvent être fournies des activités variant de 30 à 10^8 désintégrations par minute. Les sources α de contrôle sont constituées par un dépôt électrolytique de Pu_{239} en couche de masse spécifique variant de 3.10^{-9} à 0.7 mg/cm^2, ce qui correspond à des activités allant de 10 jusqu'à 10^8 désintégrations par minute.

Les sources β modèles sont constituées de radionuclides choisis de façon à assurer une gamme d'énergies limites allant de 0.018 MeV à 2.3 MeV.

Les sources β de contrôle sont faites soit de Sr_{90} + Y_{90}, soit de Tl_{204}, soit enfin de Pm_{147}. Ces sources couvrent une large gamme d'activités, entre 20 et 10^8 désintégrations par minute.

Les sources γ modèles sont constituées soit de Ra, soit de Co_{60}, soit enfin de Cs_{137}. La gamme des activités de ces étalons est comprise entre les ordres de grandeur 10^{-11} et 5 équivalents-gramme de Ra.

Les sources γ de contrôle sont préparées à partir des mêmes radionuclides que ci-dessus. Les activités couvrent une gamme très étendue, les sources ayant des formes et dimensions très variées.

Enfin, les sources modèles de neutrons sont de deux sortes: Ra-a-Be (contenant de 1 à 1000 mg de Ra) et Pu-a-Be (contenant de 0.01 à 15 g Pu). Les sources Ra-a-Be sont constituées par un mélange comprimé de RaB_2 et de Be en poudre avec un rapport en poids Ra/Be = 1/6. Les sources Pu-a-Be sont formées d'un alliage (PuBe_{91}) de ces deux métaux.

Методы метрологии радиоактивности в СССР

Авторы описывают методы, применяемые в СССР, и, в частности, в институте метрологии им. Менделеева (Ленинград) для воспроизводства при помощи стандартных аппаратов единиц измерения, используемых в радиоактивности (кури, грамм-эквивалент радия, рентген, рад).

Для радионуклидов в Советском Союзе производятся источники-еталоны двух видов:

1. Модельные источники для сравнительного эталонирования других радиоактивных препаратов, а также для калибровки радиометрических приборов и контрольных приборов за излучением;

2. Контрольные источники, служащие исключительно для проверки работы и контроля за воспроизводимостью указаний измерительных приборов.

Модельные α-источники приготавляются при помощи электролитического осаждения на платине должным образом выбранных излучателей (естественный уран, уран-233, плутоний-239, америций-241). Эти источники могут обеспечивать активность от до 10^8 распадов в минуту. Контрольные
a-источники приготавливаются путем электролитического осаждения плутония-239 в виде слоя с плотностью от $3 \times 10^{-9}$ до 0.7 мг/см², что соответствует активности от 10 до $10^8$ распадов в минуту.

Модельные β-источники приготавливаются из радионуклидов, выбранных таким образом, чтобы обеспечить диапазон энергий от 0,018 мэв до 2,3 мэв.

Контрольные β-источники приготавливаются либо из стронция-90 плюс иттрий-90, либо из таллия-204, либо из прометия-147. Эти источники охватывают широкий диапазон активности от 20 до $10^9$ распадов в минуту.

Модельные γ-источники приготавливаются либо из радия, либо из кобальта-60, либо из цезия-137. Диапазон активностей этих эталонов охватывает величины от $10^{-11}$ до 5 граммов-эквивалентов радия.

Контрольные γ-источники приготавливаются из вышеуказанных радионуклидов. Активности охватывают очень широкий диапазон, поскольку источники имеют весьма различную форму и размеры.

Наконец, модельные источники нейтронов бывают двух видов: радий-α-бериллий (содержащий от 1 до 1000 мг радия) и плутоний-α-бериллий (содержащий от 0,01 до 15 граммов плутония). Источники радий-α-бериллий приготавливаются из прессованной смеси RaBr₂ и порошкового бериллия с весовым соотношением Ra/Be = 1/6. Источники плутоний-α-бериллий образуются из сплава (PuBe₁₃) этих двух металлов.

Metodos de Metrología de la Radiactividad Aplicados en la Unión de Republicas Socialistas Sovieticas

Los autores estudian los procedimientos que se emplean en la Unión Soviética, y especialmente en el Instituto de Metrología "Mendeleev" de Leningrado, para la determinación — con ayuda de aparatos normalizados — de las unidades de medida utilizadas en la esfera de la radiactividad (curie, equivalente gramo del Ra, roentgen y rad).

En la Unión Soviética se producen actualmente dos clases de fuentes patrón:

1º Fuentes modelo que sirven para llevar a cabo trabajos de contraste por comparación de otros preparados radiactivos, así como para el calibrado de aparatos radiométricos y de monitores de radiaciones;

2º Fuentes de control, destinadas exclusivamente a la verificación del funcionamiento de los aparatos de medición y al control de la reproducibilidad de las indicaciones facilitadas por los mismos.

Las fuentes α modelo se preparan depositando electrolíticamente emisores adecuados (U natural, U²³⁸, Pu²³⁹, Am²⁴¹) sobre platino. Pueden suministrarse fuentes cuyas actividades se encuentren comprendidas entre 30 y $10^6$ desintegraciones por minuto. En cuanto a las fuentes α de control, se obtienen mediante depósito electrolítico de Pu²³⁹ en una capa cuya masa específica abarca de $3 \times 10^{-9}$ a 0.7 мг/см²; estos valores corresponden a actividades de 10 y $10^8$ desintegraciones por minuto, respectivamente.

Para las fuentes β modelo, se eligen radionúclidos que proporcionan una gama de energías límite de 0,018 a 2,3 MeV.

Las fuentes β de control están constituidas por Sr⁹⁰ + Y⁹⁰, Tl²⁰⁴ o Pm¹⁹⁷. Las actividades de estas fuentes abarcan de 20 a $10^9$ desintegraciones por minuto.
Las fuentes γ modelo están formadas por Ra, Co\(^{60}\) o Cs\(^{137}\). Los límites inferior y superior de la actividad de estos patrones son \(10^{-13}\) y 5 equivalentes gramo del Ra, respectivamente.

Las fuentes de control se preparan partiendo de los mismos radio- nucléidos que las fuentes γ modelo. Sus actividades abarcan un intervalo muy amplio y las fuentes presentan formas y dimensiones sumamente diversas.

Por último, se utilizan dos clases de fuentes neutrónicas modelo Ra-\(\alpha\)-Be (con un contenido de 1 a 1000 mg de Ra) y Pu-\(\alpha\)-Be (conteniendo de 0.01 a 15 g de Pu). Las fuentes Ra-\(\alpha\)-Be consisten en una mezcla comprimida de RaBr\(_2\) y de Be en polvo, siendo la razón ponderal Ra/Be igual a \(1/8\). En cuanto a las fuentes Pu-\(\alpha\)-Be, se preparan con una aleación (PuBe\(_{18}\)) de estos dos metales.

Основная задача радиометрии заключается в измерениях активности радиоактивных препаратов. Задачи практического использования изотопов, помимо решения этой основной задачи, требуют развития метрологических методов нахождения всех дозиметрических характеристик препаратов. Эти последние определяются: составом спектра излучения, активностью и
такими параметрами измеряемых препаратов, как размеры, химический состав, вес и данные об оболочке.

Активность препарата и спектр излучения изотопа определяют полное излучение препарата; это последнее будет отличаться от важного для практики внешнего излучения из-за поглощения и рассеяния излучения в препарате и в его оболочке. По этой причине измерения активности по-существу неотделимы от измерений интенсивности излучений, и очень часто препарат не может быть использован без знания внешнего излучения.

Комплекс необходимых метрологических мероприятий сводится к следующему:

а) унификация единиц измерений,
б) создание эталонов или эталонных методов, воспроизводящих единицы,
в) разработка системы и методов градуировки аппаратуры и паспортизации препаратов,
г) создание образцовых мер и образцовых измерительных установок, служащих для передачи размера единицы от эталонов к потребителям.

В СССР официально приняты следующие единицы: кюри, миллиграмм-эквивалент радия, рентген, рад.

Рисунок 1 б
Воспроизведение единицы кюри осуществлено в СССР с помощью следующих эталонных методов измерений и эталонных установок: 4 л-счетчиков, калориметров и ионизационных камер.

4 л-счетчики применяются для измерений α- и β-активных препаратов активностью от 10⁻⁷ до 10⁻¹¹ кюри; точность измерений 1—3% W.

Активность препаратов, распадающихся путем e-захвата (2), определяется по разности числа отсчетов 4 л-счетчика при наполнении его металом, когда регистрируются только электроны и фотony, и при добавлении в счетчик ксенона для регистрации характеристического излучения. Пределы измерения e-захватных активностей — от 10⁻¹⁰ до 10⁻⁶ кюри, точность 6—8%. 

На рисуках 1 и 2 приводится общий вид эталонных установок с 4 л-счетчиками для α- и β-частиц.

Для измерения активности препаратов по α-, β- и γ-излучению в области выше 0,01—0,05 кюри применяются следующие эталонные калориметрические установки:

а) дифференциальные γ-калориметры,
б) изотермические β- и γ-калориметры,
в) дифференциальный α-калориметр.

Установка с дифференциальными γ-калориметрами (3) (рисунок 3) содержит четыре независимых калориметра, отличающихся один от другого своими размерами (диаметры шаров 3,5; 5,5; 7 и 10 см). Каждый калориметр состоит из двух одинаковых свинцовых шаров, укрепленных в медном
боке-термостате. В центре каждого из шаров имеется полость для измеряемого препарата и электрического нагревателя. Равновесное значение разности температур шаров, измеряемого при помощи термобатареи, зависит только от тепловой мощности, выделяемой препаратом; калориметр предварительно градуируется с помощью электрического нагревателя. На калориметрах этого типа возможны измерения активностей в пределах от 0,05 до 3 кюри с точностью 3—5%. 

Изотермический β-калориметр основан на принципе испарения жидкого азота (4). Измеряемый препарат помещается в сосуд Дьюара, содержащий жидкий азот. Объем газообразного азота, выделяющийся в единицу времени, является мерой активности препарата. γ-калориметр того-же типа отличается от β-калориметра наличием специального поглотителя из вольфрама и ртути и большими размерами сосудов Дьюара. На калориметрах этого типа возможны измерения активности выше 0,01 кюри с точностью 3—6%. 

Дифференциальный α-калориметр (5) (рисунок 4), предназначен для измерения активности препаратов по α-излучению и состоит из двух одинаковых серединных стаканчиков, помещенных внутри медного блока-термостата. Чувствительность калориметра достаточна для измерения препаратов радия, содержащих не менее 1 мг радия-элемента, с точностью 1—2%. 

Ионизационный метод воспроизведения активности единицы основан на измерении ионизации, производимой γ-излучением препарата в ионизационной камере. При этом должна быть известна эффективность камеры, т. е. ионизационный ток, отнесенный к единице активности данного радиоактивного изотопа. 

Созданная щелевая ионизационная камера с телесным углом 4π (6), состоит из двух концентрически расположенных алюминиевых шаров с
наружными диаметрами 230 и 260 мм и со стенками толщиной 5 мм. Внутренний шар является собирающим электродом и соединяется с электрометрическим устройством, с помощью которого измеряется ионизация в пространстве между шарами. Измеряемый препарат помещается в центре внутреннего шара.

Рисунок 4

Экспериментально определенные и расчетные значения эффективности хорошо совпадают между собою. Расчет эффективности требует знания энергий квантов и их числа на акт распада для $\gamma$ спектров измеряемых изотопов, а также величины работы ионизации в воздухе для $\gamma$-лучей. Эта величина известна в настоящее время с погрешностью $\sim 5\%$, вследствие чего погрешность определения данным методом равна 7—8%. Пределы измерений — от $10^{-4}$ до 10 кюри.

Внешнее $\gamma$-излучение радиоактивных источников в настоящее время часто характеризуется величиной радиевого $\gamma$-эквивалента. Эта величина соответствует количеству радия (заключенного в платиновую оболочку толщиной 0,5 мм), $\gamma$-излучение которого создает в воздушно-эквивалентной камере такую же ионизацию, как $\gamma$-излучение данного источника.

Воспроизведение единицы радиевого $\gamma$-эквивалента (миллиграмм эквивалент радия) осуществляется государственным этalonом радия. До настоящего времени государственным этalonом радия СССР служил этalon № XI, сравненный со старым первичным международным этalonом радия (1911 г). Этalon № X, изготовленный и измеренный одновременно с этalonом № XI, служил этalonом-копией. В настоящее время ведутся работы по переходу на новый этalon № 5427, из числа двадцати препаратов, изготовленных Хеннигслидом в 1934 г.

Передача единицы радиевого $\gamma$-эквивалента от эталона к образцовым
препаратам радия осуществляется на эталонных установках с ионизационными камерами.

Установка для измерения в диапазоне 1—1000 мг-экв. радия (7) состоит из ионизационной камеры, соединенной с компенсационной электрометрической схемой по Таунсенду (рисунок 5). Ионизационная камера по конструкции подобна описанной выше камере для измерений активности по

**Рисунок 5**

γ-излучению, и состоит из двух концентрических шаров с наружными диаметрами 160 и 240 мм и стенками толщиной 20 мм. Точность результатов измерений составляет от 0,5% для препаратов с γ-эквивалентами ~ 10 мг-экв. радия и до 1,5% для препаратов с γ эквивалентом ~ 1000 мг-экв. радия. Препараты с γ-эквивалентом от 1 до 0,001 мг-экв. радия, создающие ионизацию, сравнимую с фоном, измеряются на установке с так называемой компенсационной камерой (6). Точность результатов лежит в пределах от 1% для препаратов в 1 мг-экв. радия до 5—8% для препаратов в 0,001 мг-экв. радия.

Воспроизведение рентгена для γ-излучения с энергией квантов от 0,3 до 3 Мэв осуществляется на эталонной установке (9) состоящей из ионизационной камеры с повышенным давлением (до 20 атм), электрометрического устройства, системы диафрагм для получения узкого пучка γ-лучей, и батареи на 15 кв.

Плоская ионизационная камера имеет потенциальный электрод длиною 75 см, измерительный электрод длиною 25 см, расположенный на расстоянии 40 см от потенциального, и два защитных электрода с двух сторон от измерительного, длиной по 30 см. Ширина всех электродов — 40 см. Измерение ионизационного тока производится компенсационным методом с помощью усилителя постоянного тока. В измеренное значение тока вводятся поправки на поглощение излучения во входном окне и в воздухе, на проникновение излучения через диафрагму, на рассеяние излучения в воздушном объеме камеры, на рекомбинацию ионов, на температуру и давление воздуха в камере.
ПОВЕРОЧНАЯ СХЕМА ДЛЯ РАДИАЦИЯРИВНЫХ ПРЕПАРАТОВ, ГРАДУИРУЕМЫХ В ЕДИНИЦАХ АКТИВНОСТИ

Эталонный метод воспроизведения единицы активности путем определения числа атомов распада

Рисунок 6
Эталонный метод воспроизведения рентгена требует знания той точки, к которой относится значение измеренного эффекта. Анализ процесса ионизации электронами показывает, что эффект должен быть отнесен к точке, несколько смещенной от центра измерительного электрода к источнику излучения (10). С учетом всех поправок точность измерений составляет 3%/6.

Помимо перечисленных эталонных работ в метрологических учреждениях СССР ведутся также определения периодов полураспада последних.

Последовательность операций для передачи размера единицы от эталона или эталонной установки к эксплуатируемой аппаратуре или рабочим препаратам описывается поверочной схемой.

В основу поверочной схемы для измерений активности положен этalonный метод воспроизведения кюри.

Следует отметить, что в СССР серийно выпускаются и применяются излучатели двух классов: образцовые, предназначенные для поверки радиоактивных препаратов и градуировки радиометрической и дозиметрической аппаратуры, а также контрольные, предназначенные исключительно для контроля работы и проверки воспроизводимости различных приборов.

Применение такого рода контрольных излучателей вызвано к жизни наличием большого числа различных дозиметрических и радиометрических приборов, используемых в стране (11) и необходимостью обеспечить контроль их работы на месте и иметь сопоставимые данные их показаний.

Препараты, изготовленные по специальным техническим условиям, и тщательно, измеренные на одной из эталонных установок, аттестуются как образцовые излучатели I разряда. Препараты, сравнимые с образцовым их излучателями I разряда, называются образцовыми излучателями II разряда и служат для градуировки дозиметрической и радиометрической аппаратуры и поверки контрольных и других рабочих излучателей.

На поверочной схеме (рисунок 6) четыре горизонтальные полосы соответствуют: эталонам и эталонным установкам, образцовым излучателям I и II разряда, рабочим препаратам. Прямоугольники изображают меры и измерительные приборы, кружки-методы измерения, измерительных установок и измеряемых препаратов. Поверочная схема устанавливает таким образом путь от любого применяемого на практике препарата или измерительного прибора к государственному эталону.

В поверочной схеме для измерений γ-эквивалентов препаратов в миллиграмм-эквивалентах радия предусмотрено сличение с помощью ионизационных камер, счетчиков. Допускается применение «скользящих», т. е. распадающихся препаратов радона.

Поверочная схема для измерений в рентгенах основана на этalonном методе воспроизведения рентгена с помощью воздушной ионизационной камеры. Передача размера единицы производится с помощью образцовых дозиметров или образцовых γ-излучателей.

При градуировке дозиметров с помощью γ-излучателей очень важно правильно выбрать расстояние и систему диафрагмирования пучка, так как эти два фактора существенно сказываются на поле излучения источника (12).

Воспроизведение единицы нейтронного потока осуществляется эталонными методами, с помощью которых калибруются образцовые нейтронные источники I-го разряда. Эти источники представляют собой препараты,
в которых бериллий облучается частицами или γ-квантами различных излучателей (радий, полоний, плутоний).

Методы определения абсолютного числа нейтронов, испускаемых нейтронными источниками, были сообщены на Второй Женевской Конференции в 1959 г. в докладе № 15/P-2087 (13). Из перечисленных в этом докладе методов практически используется в настоящее время, главным образом, метод «сопутствующих частиц» дополнительного поглотителя и наведенной β-активности.

α-излучатели

1. Образцовые α-излучатели служат для определения счетных коэффициентов α-установок, используемых для поверки α-препаратов. По этим коэффициентам определяется абсолютная α-активность поверяемых препаратов. Ввиду этого активность образцовых α-излучателей выражается числом α-распадов в телесном угле 4π без поправки на отражение частиц от подложки. При этом самопоглощение и рассеяние α-частиц в активном слое должны или отсутствовать или быть очень незначительными. Эти же α-источники могут применяться также в качестве стандартных источников тока для калибровки ионизационных камер.

Ввиду того, что счетные характеристики радиометрической аппаратуры зависят от энергии излучения, имеется необходимость изготовлять образцовые α-излучатели из различных радиоизотопов. Для изготовления образцовых α-излучателей используются U^{233}, Uр естественный, Pu^{239} и Am^{241}.

Активное вещество наносится на платиновый диск диаметром 15 мм электролитическим способом, в виде пятна диаметром 10 мм.

В таблице I указаны основные типы серийно-изготавливаемых образцовых α-излучателей (см. табл. № 1).

Из таблицы I видно, что не все типы α-излучателей выпускаются с полным диапазоном шкальности (клетки, не отмеченные знаком «+»). Это объясняется тем, что из-за значительного поглощения α-частиц в собственном слое α-излучателя и сравнительно невысокой удельной активности некоторых из них, не представляется возможным изготовить все источники с достаточно тонким слоем.

Электролитическое изготовление α-источников обеспечивает получение прочных, равномерных, хорошо сцепленных с подложкой тонких слоев. Величина интенсивности образцовых α-источников может отклоняться от номинальных значений не более чем на ±10%.

Сравнение образцовых излучателей I-го и II-го разрядов осуществляется с помощью импульсной ионизационной камеры с электронным собиранием. Активности до 30.000 распадов в минуту измеряются в ионизационной камере со значением телесного угла 4π или 2π. Активности от 30.000 и выше измеряются при малых значениях телесного угла (метод α-пушки). Точность при измерениях с геометрией 4π или 2π должна быть 0,5—1%. Каждый образцовый α-источник снабжается паспортом, где указаны: активность (выраженная числом распадов в минуту), изотопный состав, количество радиоактивных примесей, данные о периоде полуразпада, принятая схема распада, поверхностная плотность и весовые количества активного вещества, размер активного пятна, материал, толщина, вес и диаметр подложки, метод и дата измерения.
ТАБЛИЦА I
Образцовые α-излучатели

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<th>№№ пп</th>
<th>Активность в расп/мин. со всей поверхности</th>
<th>$^{233}$U</th>
<th>$^{235}$U</th>
<th>уран естественный</th>
<th>$^{239}$Pu</th>
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2. Контрольные излучатели. Активность контрольных α-излучателей выражается, как правило, числом α-распадов в минуту. Иногда активность выражается числом α-частиц в угле 2м ради 4м с поправкой на отражение частиц от подложки и на поглощение их в активном слое.

Все контрольные α-источники в СССР (за исключением источников, изготавливаемых по специальным требованиям), изготавливаются из плутония.

$^{239}$Pu по своим свойствам наиболее полно, почти идеально отвечает требованиям, которые предъявляются к радиоактивным изотопам, применяемым для изготовления образцовых и контрольных излучателей.

Нанесение плутония на подложку производится чаще всего электролитическим способом, который, благодаря высокой удельной активности плутония (0,06 кюри/г), дает достаточно тонкий (от $3 \times 10^{-9}$ до 0,7 мг/см$^2$) и вместе с тем прочно скрепленный с подложкой слой двуокиси плутония.

Для всех изготавливаемых контрольных α-излучателей установлены определенные сроки годности, зависящие от величины активности источника и условий эксплуатации. Для источников большей активности срок годности установлен больший по сравнению со слабыми источниками. Это связано с тем, что незначительные радиоактивные загрязнения таких источников будут меньше сказываться на точность измерений. Источники, применяемые в лабораторных условиях, могут служить дольше, чем такие же источники, применяемые в полевых и производственных условиях.

Ввиду того, что в настоящее время проводится много работ по измерению малых уровней радиоактивности природных объектов, в достаточно большом количестве изготавливаются комплекты источников небольшой активности на подложках малого размера для градуировки соответствующей аппаратуры.

В таблице II приведен перечень серийно выпускаемых в настоящее время источников, различной активности (от 10 до $10^8$ распадов в минуту), которые нашли широкое применение для контроля работы и различных типов дозиметрической аппаратуры.
ТАБЛИЦА II

Перечень
серийно-выпускаемых контрольных α-излучателей (по активности,
выраженной числом α-распадов в минуту со всей поверхности излучателя)

<table>
<thead>
<tr>
<th>Серия</th>
<th>Активность на поверхности</th>
<th>Активность в минуту</th>
<th>Активность в часах</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>1000</td>
<td>10000</td>
<td>100000</td>
</tr>
<tr>
<td>20</td>
<td>2000</td>
<td>20000</td>
<td>200000</td>
</tr>
<tr>
<td>40</td>
<td>4000</td>
<td>40000</td>
<td>400000</td>
</tr>
<tr>
<td>60</td>
<td>6000</td>
<td>60000</td>
<td>600000</td>
</tr>
<tr>
<td>80</td>
<td>8000</td>
<td>80000</td>
<td>800000</td>
</tr>
<tr>
<td>100</td>
<td>10000</td>
<td>100000</td>
<td>1000000</td>
</tr>
<tr>
<td>200</td>
<td>20000</td>
<td>200000</td>
<td>2000000</td>
</tr>
<tr>
<td>400</td>
<td>40000</td>
<td>400000</td>
<td>4000000</td>
</tr>
<tr>
<td>600</td>
<td>60000</td>
<td>600000</td>
<td>6000000</td>
</tr>
<tr>
<td>800</td>
<td>80000</td>
<td>800000</td>
<td>8000000</td>
</tr>
</tbody>
</table>

Всего изготавливается 20 типов комплектов α-излучателей. Входящие в
комплекты излучатели имеют различную активную поверхность (от 1 до
200 кв. см.), с различными (12 видов) подложками.
Помимо указанных в таблице I значений активности, выпускаются в
заказном порядке излучатели и с промежуточными значениями активности.

β-излучатели

1. Образцовые β-излучатели изготавливаются серией из различных
изотопов, с тем, чтобы перекрыть широкий интервал энергий, т. к. эффециентивность счетных устройств существенно зависит от энергии исследуемого
излучения. Для этих целей выбираются изотопы со сравнительно большим
периодом полураспада и как правило, — с простым спектром.
В таблице III приводится перечень изотопов, из которых делаются
образцовые излучатели, в порядке возрастания энергии β-спектра.

ТАБЛИЦА III

<table>
<thead>
<tr>
<th>Изотоп</th>
<th>Энергия, Мэв</th>
</tr>
</thead>
<tbody>
<tr>
<td>H³</td>
<td>0,018</td>
</tr>
<tr>
<td>C¹⁴</td>
<td>0,155</td>
</tr>
<tr>
<td>S³⁵</td>
<td>0,167</td>
</tr>
<tr>
<td>Cd⁴⁵</td>
<td>0,255</td>
</tr>
<tr>
<td>Co⁶⁰</td>
<td>0,31</td>
</tr>
<tr>
<td>W¹⁸⁵</td>
<td>0,43</td>
</tr>
<tr>
<td>Tl⁵²</td>
<td>0,77</td>
</tr>
<tr>
<td>Радий (D + E)</td>
<td>1,17</td>
</tr>
<tr>
<td>P⁴²</td>
<td>1,704</td>
</tr>
<tr>
<td>Sr⁹⁰ + Y⁹⁰</td>
<td>2,2</td>
</tr>
<tr>
<td>UX₁ + UX₂</td>
<td>2,3</td>
</tr>
</tbody>
</table>
Можно отметить, что в последнее время для этих целей стал использовать также прометий. Такие излучатели изготавливаются с различной активностью в диапазоне от $10^3$ до $10^6$ распад/мин.

При этом излучатели I разряда выполняются с практически невесомым слоем, нанесенным на тонкую пленку, для измерения их с помощью 4-х-счетчика. Образцовые $\beta$-излучатели II разряда готовятся нанесением радиоактивного препарата тонким равномерным слоем на подложку (обычно алюминиевую), с диаметром активного пятна в 10 мм, причем такие образцы выполняются механически прочными, химически устойчивыми, негигроскопичными и предохраняются от возможности изотопного обмена с внешней средой.

При этом радиоактивные примеси не превышают 0,5% по активности в течение срока годности образцового излучателя. Точность паспортных значений для указанных излучателей лежит в пределах от 2 до 7% в зависимости от энергии $\beta$-излучения и номинала активности.

2. Контрольные $\beta$-излучатели изготавливаются в основном из Sr$^{90}$, находящегося в равновесии с Y$^{90}$, частично из Th$^{224}$ и Pm$^{147}$. Активность контрольных излучателей выражается числом $\beta$-распадов в минуту. Изготавливаются источники либо закреплением соли Sr$^{90}$ непосредственно на подложке, либо на основе фенолформальдегидной смолы или наполнением оксидной пленки на алюминии. Прометий и таллий наносятся на подложку электрохимическим путем. Изготовленные различными способами $\beta$-излучатели обеспечивают достаточную равномерность нанесения активного вещества на подложке и надежность излучателя при эксплуатации в различных условиях. Некоторые источники заклеиваются алюминиевой фольгой для предохранения активного слоя от механического воздействия.

Контрольные $\beta$-источники изготавливаются в широком диапазоне активности, от 20 до 10$^9$ распадов в минуту, что соответствует удельной поверхностью активности на один кв. см. от 0,2 до 2.10$^8$ распадов в минуту. Шкала активности $\beta$-излучателей установлена такая же, как и для контрольных $\alpha$-излучателей. Так же, как и для $\alpha$-излучателей, в заказном порядке выпускаются $\beta$-излучатели с промежуточными значениями активности.

Изготавливается 25 различных комплектов контрольных $\beta$-излучателей на разных подложках 13-ти типов. Эти излучатели имеют активную поверхность от 0,1 до 300 кв. см.

Контрольные $\beta$-излучатели снабжаются паспортом, где указывается: активность в распадах в минуту, точность и дата измерения, неравномерность нанесения активного слоя и т. д.

Неравномерность нанесения активного слоя определяется как отклонение в процентах контрольного измерения в любой точке активной поверхности излучателя от среднед-арифметического значения активности одного квадратного сантиметра поверхности излучателя.
Неравномерность проверяется путем измерения части активной поверхности источника в один кв. см. в нескольких местах (от 3 до 12, в зависимости от геометрических размеров излучателя) посредством соответствующего диафрагмирования активной поверхности.

При установлении срока годности для различных контрольных \(\beta\)-излучателей руководствуются помимо соображений, указанных для \(\alpha\)-излучателей, еще и конструктивным оформлением препарата, т. е. защищен ли он прочной оболочкой, иными словами является ли он закрытым или открытым источником.

\(\gamma\)-излучатели

1. Образцовые \(\gamma\)-излучатели изготавливаются из радиоизотопов с коэффициентами, указанными для \(\alpha\)-излучателей, в зависимости от геометрических размеров излучателя (в гр. экв. радия): 5; 2, 1; 0,500; 0,200; 0,100; 0,025; 0,015; 0,005; 0,001; 10\(^{-4}\); 10\(^{-5}\); 10\(^{-6}\); 10\(^{-7}\); 10\(^{-8}\); 10\(^{-9}\); 10\(^{-10}\); 10\(^{-11}\).

Величина радиевого \(\gamma\)-эквивалента может отличаться от номинала не более, чем на \(\pm 20\%\).

Образцовые \(\gamma\)-излучатели из \(\mathrm{Co}^{60}\) изготавливаются в виде шариков или цилиндриков размерами 1—2 мм. Каждый излучатель заключается в герметичную алюминиевую оболочку со стенками толщиной 0,2 мм.

Образцовые \(\gamma\)-излучатели из \(\mathrm{Cs}^{137}\) (1 мг. экв. радиоизотопа и ниже) представляют собой ампулу цилиндрической формы, внутренней стенкой которой служит \(\mathrm{Cs}^{137}\), нанесенный на шарик диаметром 0,25—1 мм. Излучатели с \(\gamma\)-эквивалентом большим 1 мг. экв. радиоизотопа выполняются в виде цилиндрических ампул из нержавеющей стали диаметром до 10 мм и длиной до 15 мм.

Образцовые излучатели из радиоизотопа (с содержанием радиоизотопа от 10\(^{-7}\) до 10\(^{-11}\) гр) представляют собой водный раствор хлористого радия, содержащий определенную примесь хлористого бария, и заключенный в цилиндрическую стеклянную ампулу.

Образцовые радиевье излучатели с содержанием радиоизотопа менее 10\(^{-7}\) грамма (жидкие) имеют следующие характеристики:

<table>
<thead>
<tr>
<th>№№</th>
<th>Номинальное содержание радия в граммах</th>
<th>Размеры ампулы в см</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>диаметр</td>
</tr>
<tr>
<td>1</td>
<td>(10^{-7})</td>
<td>от 0,6 до 1,5</td>
</tr>
<tr>
<td>2</td>
<td>(10^{-8})</td>
<td>&quot;</td>
</tr>
<tr>
<td>3</td>
<td>(10^{-9})</td>
<td>&quot;</td>
</tr>
<tr>
<td>4</td>
<td>(10^{-10})</td>
<td>&quot;</td>
</tr>
<tr>
<td>5</td>
<td>(5 \cdot 10^{-11})</td>
<td>&quot;</td>
</tr>
</tbody>
</table>

ПРИМЕЧАНИЕ: п—любое число от 1 до 9, либо определенное, т. е. заранее оговоренное.

Содержание радиоизотопа в готовом препарате определяется изготовителем расчетным путем по навеске исходного раствора и концентрации его. Исходный раствор, промерен с точностью до 0,7%. Паспорт на излучатель содержит следующие данные: содержание радиоизотопа в граммах, дата
изготовления, содержание бария, данные (диаметр, длина, толщина стенок).
Образцовые излучатели из радия, с содержанием от 0,2 до $10^{-6}$ г. представляют собой бромистую соль радия, помещенную в стеклянную ампулу. Ампула дополнительно заключается в футляр, изготовленный из сплава (90% платины и 10% иридия), с толщиной стенки 0,5 мм. Краткая характеристика изготавливаемых г-излучателей из радия приведена в таблице V. Ампулы изготавливаются из безборного стекла марки Ц-32.

ТАБЛИЦА V
Характеристика радиевы г-излучателей

<table>
<thead>
<tr>
<th>№ п/п</th>
<th>Содержание радия в гр</th>
<th>Допустимое отклонение по содержанию радия в %</th>
<th>Размеры наружной стеклянной ампулы в мм</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0,2</td>
<td>± 5</td>
<td>6,9—7,8</td>
</tr>
<tr>
<td>2</td>
<td>0,1</td>
<td>&quot;</td>
<td>&quot;</td>
</tr>
<tr>
<td>3</td>
<td>0,025</td>
<td>± 10</td>
<td>&quot;</td>
</tr>
<tr>
<td>4</td>
<td>0,010</td>
<td>&quot;</td>
<td>&quot;</td>
</tr>
<tr>
<td>5</td>
<td>0,005</td>
<td>&quot;</td>
<td>&quot;</td>
</tr>
<tr>
<td>6</td>
<td>0,001</td>
<td>&quot;</td>
<td>&quot;</td>
</tr>
<tr>
<td>7</td>
<td>$10^{-4}$</td>
<td>± 20</td>
<td>&quot;</td>
</tr>
<tr>
<td>8</td>
<td>$10^{-5}$</td>
<td>&quot;</td>
<td>&quot;</td>
</tr>
<tr>
<td>9</td>
<td>$10^{-6}$</td>
<td>&quot;</td>
<td>&quot;</td>
</tr>
</tbody>
</table>

В паспорте на образцовый г-излучатель указывается: вес безводной бромистой соли радия в гр, содержание радия в источнике в гр, точность определения, дата измерения.

2. Контрольные г-излучатели изготавливаются из тех же изотопов, что и образцовые, в широком диапазоне активностей.
Помимо упомянутых форм, контрольные излучатели из Co$^{60}$ выпускаются в виде цилиндров размером 1 × 1 мм, 2 × 2 мм и 5 × 5 мм (в алюминиевой оболочке) и в виде отрезков проволоки диаметром в
0,7—0,9 мм; излучатели из Cs\(^{137}\) выпускаются тех же типов, что и образцовые. Кроме того выпускается большое число контрольных \(\gamma\)-излучателей самых разнообразных форм и размеров, применительно к специализированным приборам. Некоторые из них изображены рисунке 7.

**Нейтронные источники**

1. Образцовые источники. В качестве образцовых нейтронных источников принят набор радий-\(\alpha\)-бериллиевых источников, содержащих: 1000, 300, 100, 30, 10 и 1 мг радия, а также набор плутоний-\(\alpha\)-бериллиевых источников, содержащих 15, 1, 0,5, 0,1 и 0,01 гр плутония.

Плутониево-бериллиевые источники представляют собой интерметаллическое соединение Pu\(^{239}\) с бериллием (PuBe\(_3\)). Радий-\(\alpha\)-бериллиевые источники представляют собой прессованную смесь бромида радия с порошкообразным бериллием, заключенную в герметическую двойную оболочку цилиндрической формы. Соотношение весовых количеств радия и бериллия в смеси должно быть 1 : 6.

Выход нейтронов на 1 мг. радия в угле 4 л составляет не менее 1 . 10\(^4\) нейтронов в секунду. Радий должен быть свободен от мезотория и не должен содержать более, чем 5\(^\circ\)/\(^\circ\) бария.

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The basic purpose of radiometry is to measure the activity of radioactive preparations. The practical use of isotopes requires, in addition to the fulfilment of this basic purpose, the development of metrological methods of finding all the dosimetric characteristics of preparations. These characteristics are determined by the composition of the radiation spectrum, the activity, such parameters of the preparations to be measured as size, chemical composition and weight, and data concerning the casing.

The activity of the preparation and the radiation spectrum of the isotope determine the total radiation of the preparation; this total radiation will be found to differ from the external radiation, which is the significant factor in practice, owing to absorption and scattering in the preparation and the casing. For this reason, the activity measurements are fundamentally inseparable from the measurements of the radiation intensity, and in many cases a preparation cannot be utilized if the external radiation is unknown.

The full set of metrological procedures required is as follows:
(a) Unification of the units of measurement;
(b) Preparation of standards, or standard methods of reproducing the units;
(c) Development of a system and methods of calibrating the apparatus and preparing specifications of the preparations; and
(d) Establishment of model measures and model measuring apparatus for use in transmitting the unit dimension from the standards to the users.

In the USSR the following units have been officially adopted: the curie, the milligramme-equivalent of radium, the roentgen and the rad.

The curie unit is reproduced in the USSR by means of the following standard methods of measurement and standard apparatus: 4π counters, calorimeters and ionization chambers.

4π counters are used for measuring α and β emitting preparations of an activity of $10^{-7} - 10^{-11}$ c; the measurement accuracy is 1–3%. The activity of preparations decaying by electron capture (2) is determined by reference to the difference in the number of counts recorded by the 4π counter when the counter is filled with methane (only electrons and background then being counted) and when xenon is added to the counter in order to count the characteristic radiation. The possible range of measurement of electron-capture activity is $10^{-10} - 10^{-8}$ c, and the accuracy is 6–8%.

Figures 1 and 2 give a general view of standard apparatus using 4π counters for α and β particles.

For the measurement of the α, β and γ activity in the region above 0.1–0.05 c, the following standard calorimetric apparatus is used:
(a) Differential γ calorimeters;
(b) Isothermal β and γ calorimeters; and
(c) Differential α calorimeters.
The set-up for the differential $\gamma$ calorimeters (3) (figure 3) consists of four independent calorimeters, differing from one another in size (the ball diameters are 3.5, 5.5, 7 and 10 cm. respectively). Each calorimeter consists of two identical lead balls fixed in a copper thermostat block. In the centre of each ball there is a cavity to accommodate the preparation to be measured and an electric heater. The equilibrium value of the difference of the temperatures of the balls, which is measured with a thermopile, depends solely on the thermal power emitted by the preparation; the calorimeter is previously calibrated by means of the electric heater. Calorimeters of this type are capable of measuring activities in the range 0.5—3 c with an accuracy of 3—5%.

The isothermal $\beta$ calorimeter works on the principle of vaporization of liquid nitrogen (4). The preparation to be measured is placed in a Dewar flask containing liquid nitrogen. The volume of gaseous nitrogen evolved per unit of time acts as a measure of the radioactivity of the preparation. The isothermal $\gamma$ calorimeter differs from the $\beta$ calorimeter in having a special tungsten and mercury absorber and larger Dewar flasks. With calorimeters of this type, activities over 0.1 c may be measured to an accuracy of 3—6%.

The differential $\alpha$ calorimeter (5) (figure 4) is designed to measure the activity of preparations by their $\alpha$ emission and consists of two identical silver cups arranged inside a copper thermostat block. This calorimeter is sensitive enough to measure radium preparations containing not less than 1 mg. of elemental radium to an accuracy of 1—2%.

The ionization method of reproducing unit activity works on the principle of measuring the ionization produced in an ionization chamber by the $\gamma$ radiation of the preparation. For this, the efficiency of the chamber, i.e. the ionization current associated with unit activity of the radioisotope in question, must be known.

The $4\pi$ solid-angle slit ionization chamber (6) which has been evolved, consists of two concentrically disposed aluminium spheres with outside diameters of 230 and 260 mm and walls 5 mm thick. The inner sphere is the collecting electrode and is connected with an electrometer which measures the ionization in the space between the spheres. The preparation to be measured is placed in the centre of the inner sphere.

There is good agreement between the values found by experiment and the calculated values for the efficiency of the apparatus. To calculate the efficiency, the quantum energies and the number of quanta per disintegration for the $\gamma$-spectra of the isotope to be measured, as well as the working ionization quantity in air for $\gamma$-rays, must be known. This quantity is at present known with an error of $\sim 5\%$, so that the error in determining the activity by this method is 7—8%. The range of measurement is $10^{-4}$—10 c.

The external $\gamma$ radiation emitted by radioactive sources is at present frequently defined in terms of the $\gamma$ equivalent of radium. This quantity is the quantity of radium in a platinum blanket 0.5 mm thick, the $\gamma$ radiation of which produces the same amount of ionization in an air-equivalent chamber as the $\gamma$ radiation of the source in question.

The unit of the radium $\gamma$ equivalent (the milligramme-equivalent of radium) is reproduced by the State Radium Standard. Up to the present, the USSR State Radium Standard has been Standard No. XI, which was compared with the old primary international radium standard (1911). Standard No. X, which was prepared and measured at the same time as Standard No. XI,
has served as a duplicate standard. Arrangements are now in hand to go over
to the new standard No. 5427 from among 12 preparations produced by
Hoenigschmidt in 1934.

The transmission of the unit of γ equivalent of radium from the standard
to the radium model preparations is carried out with standard apparatus
incorporating ionization chambers.

The apparatus for making measurements in the 1—1000 mg Ra range (7)
consists of an ionization chamber in association with a Townsend compensation
electrometric circuit (figure 5). The ionization chamber is similar in design to
the one used for measurement of γ activity described above; it consists of
two concentric spheres with outside diameters of 160 and 240 mm. and walls
20 mm. thick. The accuracy of the measurement results attained with this
apparatus varies from 0.5% in the case of preparations with a γ equivalent
of ~ 10 mg Ra to 1.5% in the case of preparations with a γ equivalent of
~ 1000 mg Ra. Preparations with a γ equivalent of to 0.001 mg Ra, emitting
radiation comparable to the background, are measured by an apparatus using
a “compensation chamber” (8). The accuracy ranges from 1% for preparations
of 1 mg Ra to 5—8% for those of 0.001 mg Ra.

The roentgen, for γ radiation with quantum energies of 0.3—3 MeV,
is reproduced on a standard apparatus (9) consisting of a high-pressure (up to
20 atm) ionization chamber, an electrometer, a system of diaphragms for
producing a narrow γ-ray beam, and a 15-kV battery.

The flat ionization chamber has a potential electrode 75 cm. long, a
measuring electrode 25 cm long set up 40 cm away from the potential electrode.
The width of all the electrodes is 40 cm. The ionization current is measured
by the compensation method, using a direct-current amplifier. The measured
reading for the current is adjusted for absorption of radiation in the entry
window and in the air, penetration through the diaphragm, scattering in the
chamber air-space, recombination of ions and air temperature and pressure in
the chamber.

The standard method of reproduction of the roentgen requires that the point
to which the value of the measured effect relates be known. Analysis of the
electron ionization process shows that the effect must be related to a point
somewhat displaced from the centre of the measuring electrode towards the
radiation source (10). After making allowance for all adjustments, the accuracy
of the measurements is 3%.

In addition to the standards work listed above, precision measurements of
the radiation spectra of various isotopes and of their half-lives are also per-
formed in the metrological centres of the USSR. The sequence of operations
for transferring the unit size from the standard or standard apparatus to the
instrument to be used or to the working preparations is set out in a control
diagram.

The standard method of reproducing the curie is taken as the basis for
this control diagram.

It should be noted that emitters of two types are in regular production
and use in the USSR: they are model emitters, designed for checking radio-
active preparations and calibrating radiometric and monitoring apparatus, and
control emitters, designed exclusively for verifying the functioning and
checking the reproducibility of various instruments.
The use of these control emitters was made necessary by the large number of different monitoring and radiometric instruments in use in the country (11) and the need to check their functioning on the spot and for their readings to be comparable.

Preparations which have been manufactured under special technical conditions and carefully measured on a standard apparatus are certified as grade I model emitters. Preparations that have been compared with grade I model emitters are termed grade II model emitters and are used for calibrating monitoring and radiometric equipment and for checking control and other working emitters.

In the control diagram (figure 6), the four horizontal bands correspond respectively to standards and standard apparatus, grade I model emitters, grade II model emitters and working preparations. The rectangles represent measures and measuring instruments and the circles represent measurement methods, the measuring apparatus and the preparations to be measured. The control diagram thus establishes the path leading from any preparation or measuring instrument used in practice to the State standard.

The control diagram for measurements of the \( \gamma \) equivalents of preparations in milligramme-equivalents of radium provides for the comparison of counters by means of ionization chambers. The use of decaying radon preparations is permitted.

The control diagram for measurements in roentgens is based on the standard method of reproducing the roentgen by means on an air ionization chamber. The unit size is transferred by means of model dosimeters or \( \gamma \) emitters.

When calibrating monitoring equipment by means of \( \gamma \) emitters, it is very important to select the distance and the beam masking system correctly, as both these factors have a substantial influence on the field of radiation of the source (12).

The unit of neutron flux is reproduced by the standard methods used to calibrate grade I model neutron sources. These sources take the form of preparations in which beryllium is irradiated by particles or \( \gamma \) quanta from various emitters (radium, polonium or plutonium). Methods of determining the absolute number of neutrons emitted by neutron sources were reported at the second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958, in paper A/CONF. 15/P/2087 (13). Of the methods enumerated in this paper, the one mainly used in practice at present is that involving "accompanying particles", a supplementary absorber and induced \( \beta \) activity.

As we have already noted, model emitters and sources account for a substantial portion of the control diagrams. A short description of the various types of emitter and source is given below.

**\( \alpha \) emitters**

1. **Model \( \alpha \) emitters** are used to determine the counting coefficients of \( \alpha \) apparatus used to check \( \alpha \) preparations. The absolute \( \alpha \) activity of the preparations to be checked is determined in accordance with these coefficients. Consequently, the activity of the model \( \alpha \) emitters is expressed by the number of \( \alpha \) disintegrations in a complete \((4 \pi)\) solid angle, with no correction for reflexion of particles from the mount. In these conditions self-absorption and scattering of \( \alpha \) particles in the active layer must be either entirely absent or
quite insignificant. These same α sources can also be used as standard current sources for calibrating ionization chambers.

Since the counting characteristics of radiometric instruments depend on the radiation energy, model α emitters must be prepared from a variety of radioisotopes. U\textsuperscript{233}, natural uranium, Pu\textsuperscript{239}, and Am\textsuperscript{241} are used for this purpose.

The active substance is applied electrolytically in the form of a spot 10 mm in diameter to a platinum disc with a diameter of 15 mm.

The main types of regularly prepared model α emitters are listed in Table I. It will be seen from the table that not all types of α emitters are supplied to cover a full range of the activity scale (the boxes not marked with "+" signs). This is because, due to the considerable absorption of α particles in the α emitting layer proper and to the comparatively low specific activity of some of the emitters, not all the sources can be prepared with a thin enough layer.

The preparation of α sources by electrolysis ensures that strong uniform thin layers firmly secured to the mount will be obtained. The value for the intensity of model α sources may not deviate from the rated values by more than ±10%.

| Table I |
| Model α emitters |

<table>
<thead>
<tr>
<th>No.</th>
<th>Activity in disintegrations/minute from whole surface</th>
<th>Uranium 233</th>
<th>Uranium 235</th>
<th>Natural uranium</th>
<th>Plutonium 239</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>30</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>2</td>
<td>100</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>3</td>
<td>300</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>4</td>
<td>1 000</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>5</td>
<td>3 000</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>6</td>
<td>10 000</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>7</td>
<td>30 000</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>8</td>
<td>100 000</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>9</td>
<td>300 000</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>10</td>
<td>1 000 000</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>11</td>
<td>3 000 000</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>12</td>
<td>10 000 000</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
</tr>
</tbody>
</table>

Grade I and grade II model emitters are compared by means of a counting ionization chamber with electron collection. Activities of up to 30,000 disintegrations per minute are measured in the ionization chamber with a solid angle of 4π or 2π. Activities of 30,000 or more are measured using small solid angles (α gun method). The accuracy of measurement using the 4π or 2π method must be 0.5—1%.

Each model α source is supplied with a “passport” or specification stating its activity (in terms of disintegrations per minute), the isotopic composition, the amount of radioactive impurities, data on the half-life, the accepted decay scheme, the superficial density and the quantity of active material by weight, the size of the active spot, the material, thickness, weight and diameter of the mount and the method and date of measurement.

2. Control emitters: The activity of control α emitters is usually expressed by the number of α disintegrations per minute. Occasionally it is
expressed by the number of $\alpha$ particles in a $2\pi$ or $4\pi$ angle with a correction for reflexion of particles from the mount and for their absorption in the active layer.

All the control $\alpha$ emitters produced in the USSR are prepared from plutonium, except for sources prepared against special orders.

The properties of Pu$^{239}$ are such that it meets more fully than any other radioisotope, indeed almost ideally, the demands made on radioisotopes to be used in preparing model and control emitters.

The plutonium is most often applied to the mount by the electrolytic method which, due to the high specific activity of plutonium (0.06 curie/g), yields a thin enough layer of plutonium dioxide (from $3 \times 10^{-9}$ to 0.7 mg/cm$^2$) which is at the same time firmly attached to the mount.

Definite periods of useful life, depending on the activity of the source and the conditions of use, have been established for all the control $\alpha$ emitters to be prepared. The useful life of high-activity sources has been set at a relatively higher figure than that of low-activity sources. This is because the insignificant radioactive contaminations of high-activity sources will have less effect on the accuracy of the measurements. Sources employed in laboratory conditions may be used for longer than sources used in field and industrial conditions.

In view of the considerable work being done at present on the measurement of low levels of radioactivity of natural objects, sets of low-activity sources on small mounts for the calibration of the necessary instruments are being prepared in adequate quantity. Table II lists the sources of various activities (from 10 to $10^8$ disintegrations per minute) at present in current production which have gone into general use for checking the functioning of various kinds of monitoring instrument.

**TABLE II**

List of Control $\alpha$ Emitters in Current Production (Activity Expressed as the Number of Disintegrations per Minute over the Whole Surface of the Emitter)

<table>
<thead>
<tr>
<th>Activity (Disintegrations per Minute)</th>
<th>Surface (cm$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>1 000</td>
</tr>
<tr>
<td>20</td>
<td>2 000</td>
</tr>
<tr>
<td>40</td>
<td>4 000</td>
</tr>
<tr>
<td>60</td>
<td>6 000</td>
</tr>
<tr>
<td>80</td>
<td>8 000</td>
</tr>
<tr>
<td>100</td>
<td>10 000</td>
</tr>
<tr>
<td>200</td>
<td>20 000</td>
</tr>
<tr>
<td>400</td>
<td>40 000</td>
</tr>
<tr>
<td>600</td>
<td>60 000</td>
</tr>
<tr>
<td>800</td>
<td>80 000</td>
</tr>
</tbody>
</table>

A total of 20 different sets of $\alpha$ emitters are produced. The emitters making up the sets have various active surfaces (from 1 to 200 sq. cm) in 12 kinds of mount.

In addition to the activities listed in Table I, emitters with intermediate levels of activity are supplied to order.

**$\beta$ emitters**

1. Model $\beta$ emitters are regularly prepared from various isotopes to cover a wide range of energies, since the efficiency of counters fundamentally depends
on the energy of the radiation under study. Isotopes with a comparatively long half-life and, as a rule, a simple spectrum are chosen for these purposes.

Table III gives a list of the isotopes from which model emitters are made in order of increasing β spectrum energy.

<table>
<thead>
<tr>
<th>Table III</th>
</tr>
</thead>
<tbody>
<tr>
<td>Isotope</td>
</tr>
<tr>
<td>H³</td>
</tr>
<tr>
<td>C¹⁴</td>
</tr>
<tr>
<td>Sr⁵⁵</td>
</tr>
<tr>
<td>Ca⁴⁵</td>
</tr>
<tr>
<td>Co⁶⁰</td>
</tr>
<tr>
<td>W⁹⁸</td>
</tr>
<tr>
<td>Tl²⁰⁴</td>
</tr>
<tr>
<td>Radium (D + E)</td>
</tr>
<tr>
<td>P³²</td>
</tr>
<tr>
<td>Sr⁹⁰ + Y⁹⁰</td>
</tr>
<tr>
<td>UX₁ + UX₂</td>
</tr>
</tbody>
</table>

It may be mentioned that promethium has recently also begun to be used for these purposes. Promethium emitters are prepared with various activities in the range $10^4$—$10^6$ disintegrations per minute.

Grade I emitters are prepared in the form of a practically weightless layer applied to a thin film, so that they can be measured with a $4\pi$ counter. The model β emitters of grade II are prepared by applying the radioactive preparation as a thin uniform layer to the mount (usually of aluminium), the active spot being 10 mm in diameter; these sources are mechanically strong, chemically stable and non-hygroscopic and are protected from possible isotope exchange with an external medium.

As in the case of the α emitters, the accepted decay scheme, and half-life, the size, composition and thickness of the active layer, data on impurities and details of the method of measurement are all specified in the "passport" or specification attached to a β source. The period of validity of these specifications is fixed at one year in the case of long-lived (over five years) sources and at one half-life (but not more than one year) in the case of short-lived emitters. At the same time, radioactive trace impurities are not to exceed 0.5% of the activity during the useful life of a model emitter. The accuracy of the data given in the specifications is 2—7%, depending upon the energy of the β radiation and the nominal activity.

2. Control β emitters are prepared mainly from Sr⁹⁰ in equilibrium with Y⁹⁰ but sometimes from Tl²⁰⁴ and Pm¹⁴⁷. The activity of the control emitters is expressed as the number of β disintegrations per minute. The sources are prepared by fixing a salt of Sr⁹⁰ either directly on the mount or on a phenol-formaldehyde resin base or by applying an oxide film to aluminium. Promethium and thallium are applied to the mount by electrochemical means. The β emitters prepared by these various methods, provide a sufficiently uniform coating of active substance on the mount and ensure reliability of the source when operating
in various conditions. Some of the sources are covered with aluminium foil to protect the active layer from mechanical influences.

Control $\beta$ sources are prepared in a wide range of activities (20 to $10^9$ disintegrations per minute) corresponding to specific surface activities of 0.2—$2 \times 10^8$ disintegrations per minute per sq. cm. The same range of activities is established for the $\beta$ emitters as for the control $\alpha$ emitters. As in the case of $\alpha$ emitters, $\beta$ emitters with intermediate levels of activity are prepared to order.

Twenty-five different sets of control $\beta$ emitters are prepared on 13 different kinds of mount. The emitters have active surfaces of 0.1—300 sq. cm.

The control emitters are supplied with a "passport" stating the activity in disintegrations per minute, the accuracy and date of measurement, the degree of non-uniformity in the application of the active layer, etc.

The degree of non-uniformity in the application of the active layer is defined as the percentage deviation of a measurement check at any point on the active surface of the emitter from the arithmetical mean activity over one square centimetre of the surface.

The non-uniformity is checked by measuring sections of the active surface of the source totalling 1 sq. cm. in several places (from 3 to 12, depending upon the geometric dimensions of the emitter) by suitably masking the active surface.

Those responsible for establishing the period of useful life of various control emitters are guided not only by the considerations — already mentioned in connexion with $\alpha$ emitters but also by the structural form taken by the preparation, — i. e., whether or not it is protected by a strong casing or, in other words, whether it is an open or a closed source.

$\gamma$ emitters

1. Model $\gamma$ emitters are prepared from radium, $\text{Co}^{60}$ and $\text{Cs}^{137}$. The series of model emitters consists of preparations having the following radium $\gamma$ equivalents (in gramme-equivalents of radium): 5, 2.1, 0.500, 0.200, 0.100, 0.025, 0.010, 0.005, 0.001, $10^{-4}$, $10^{-5}$, $10^{-6}$, $10^{-7}$, $10^{-8}$, $10^{-9}$, $10^{-10}$ and $10^{-11}$. The value of the radium $\gamma$ equivalent may vary from the rated value by not more than 20% either way.

Model $\gamma$ emitters of $\text{Co}^{60}$ are prepared as small pellets or cylinders 1—2 mm in diameter. Every emitter is enclosed in an airtight aluminium casing with walls 0.2 mm thick.

Model $\gamma$ emitters of $\text{Cs}^{137}$ (1 mg radium equivalent or less) take the form of spherical stainless-steel ampoules with walls 1.2 mm thick, the $\text{Cs}^{137}$ being applied to a pellet 0.25 to 1 mm in diameter which is then placed in the ampoule. Emitters having a $\gamma$ equivalent greater than 1 mg Ra take the form of cylindrical stainless-steel ampoules 10 mm in diameter and 15 mm long.

Model emitters prepared from radium (with a radium content of $10^{-7}$—$10^{-11}$ g) take the form of an aqueous solution of radium chloride with a measured addition of barium chloride, the whole being placed in a cylindrical glass ampoule.

Model radium emitters with a radium content of less than $10^{-7}$ g (liquid) have the following features:
TABLE IV

<table>
<thead>
<tr>
<th>Ampoule dimensions in cm</th>
<th>Rated contents of radium in grammes</th>
</tr>
</thead>
<tbody>
<tr>
<td>diameter</td>
<td>length</td>
</tr>
<tr>
<td>from 0.6 to 1.5</td>
<td>5—12</td>
</tr>
<tr>
<td>&quot;</td>
<td>&quot;</td>
</tr>
<tr>
<td>&quot;</td>
<td>&quot;</td>
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<td>&quot;</td>
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<tr>
<td>&quot;</td>
<td>&quot;</td>
</tr>
</tbody>
</table>

Note: n = any numbers from 1 to 9 or a specific number previously agreed.

The radium content of the finished preparation is determined by the manufacturer by calculation on the basis of the weight and concentration of the starting solution. The starting solution is measured to an accuracy of 0.7%. The specification accompanying the emitter contains the following information: the content of elemental radium in grammes, the date of preparation, the barium content and details of the ampoule (diameter, length and wall thickness).

The model emitters with a radium content of $0.2 - 10^{-6}$ g take the form of a bromide salt of radium in a glass ampoule. The ampoule is further enclosed in a case made from a platinum-iridium alloy (90% platinum and 10% iridium) with walls 0.5 mm thick. A brief specification of the radium \( \gamma \) emitters prepared are given in Table V. The ampoules are made from TS-32 boron-free glass.

TABLE V

<table>
<thead>
<tr>
<th>Specification of Radium ( \gamma ) Emitters</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radium content in g</td>
</tr>
<tr>
<td>---------------------</td>
</tr>
<tr>
<td>1</td>
</tr>
<tr>
<td>2</td>
</tr>
<tr>
<td>3</td>
</tr>
<tr>
<td>4</td>
</tr>
<tr>
<td>5</td>
</tr>
<tr>
<td>6</td>
</tr>
<tr>
<td>7</td>
</tr>
<tr>
<td>8</td>
</tr>
<tr>
<td>9</td>
</tr>
</tbody>
</table>

The specification for model \( \gamma \) emitters indicates the weight of the anhydrous bromide salt of radium in g, the radium content of the source in g, the accuracy of determination and the date of measurement.

2. Control \( \gamma \) emitters are prepared from the same isotopes as the model emitters over a wide range of activities.

In addition to the forms already mentioned, Co$^{60}$ control emitters are prepared in the form of cylinders $1 \times 1$, $2 \times 2$ and $5 \times 5$ mm (in an aluminium casing) and in the form of pieces of wire 0.7—0.9 mm in diameter; the Cs$^{137}$
control emitters are prepared in the same forms as the model of shapes and sizes are supplied for specialized instruments. Some of these are caesium emitters. Also, a large number of control γ emitters in the same range is illustrated in figure 7.

**Neutron sources**

1. **Model sources.** A series of radium-α-beryllium sources containing 1.000, 300, 100, 30, 10 and 1 mg of radium and a series of plutonium-α-beryllium sources containing 15, 1, 0.5, 0.1 and 0.01 g of plutonium are used as model neutron sources.

The plutonium-beryllium sources take the form of an intermetallic union of Pu and beryllium (PuBe).

The radium-α-beryllium sources are in the form of a compressed mixture of radium bromide and powdered beryllium in a gastight, double-walled cylindrical casing. The proportion of radium to beryllium in the mixture must be $1:6$ by weight.

The neutron production per 1 mg of radium in a $4\pi$ angle is not less than $1 \times 10^4$ neutrons per second. The radium must be free of mesothorium and must not contain more than 5% of barium.

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I propose just to give you a very brief tour through the Radioactivity Section of the National Bureau of Standards. In so doing I will not, however, refer to the preparation and calibration of neutron standards since this is the responsibility of the Neutron Physics Section at the NBS. Figure 1 shows the certificate for the first radioactivity standards at the National Bureau of Standards; that for the radium standard that Madame Curie brought to the United States in 1913 is on the left, and the certificate for one of our 1934 Hönigschmid standards is on the right. It is interesting to note the family continuity over a period of nearly 25 years. Stefan Meyer signed both certificates, Mme. Joliot-Curie had succeeded her mother on the International Radium Standards Commission and Rutherford having become a baron had been able to dispense with his initial. As there was only one Rutherford one might say he did not really need an initial.

Radium standards are standards by weight, but they can equally well be standardized by measuring their rate of energy emission. At the National Bureau of Standards we have built a microcalorimeter which is shown diagrammatically in figure 2. This calorimeter was actually built to accommodate
CERTIFICAT.

La Présidente de la Commission de Radiométrie de l'Union internationale des radiotélégraphistes a examiné, le 25 septembre 1924, l'émission des appareils de la Société SNCF, et a constaté que l'émission correspondait exactement à la specification.

La présente est signée par la Présidente de la Commission de Radiométrie de l'Union internationale des radiotélégraphistes.

Société SNCF.

Figure 1

Figure 2

90
our Hönigschmid standards for intercomparison with other national radium standards. There are thermocouples around the equator of each cup and the cold junctions dip into silicone oil in cavities in the bottom copper block; silicone oil was chosen because of its low vapor pressure. On the bottom of each cup there is soldered a Peltier couple by means of which you can balance the energy being emitted by the radioactive source.

The next figure shows a photograph of the actual microcalorimeter, which is supported, in use, inside a temperature-attenuating enclosure. Because the design of this microcalorimeter is based on the principle of compensation it is possible to take readings about half an hour after starting a run, and there is no appreciable waiting period. Consequently there is no need for a thermostatically-controlled enclosure; a temperature-attenuating enclosure is sufficient.

Figure 4 shows some of the results which have been obtained at the National Bureau of Standards comparing our own with other Hönigschmid radium standards, both with the gold-leaf electroscope and with the microcalorimeter and also some measurements carried out at the National Physical Laboratory in which one of our standards was compared with the British standard. It is rather interesting to compare the electroscope ratios with the microcalorimeter ratios. These ratios have always been shown in such a way that the numerator is larger than the denominator. In every case, except one, the ratio is higher for the microcalorimeter than for our measurements with the electroscope, but this discrepancy is only slightly greater than the precision of about 0.1%, so that it is a very marginal effect. But in practically every case, I think, these values are slightly outside the experimental uncertainty and show the effect of γ-ray self-absorption which becomes apparent in the normal way of measuring with an electroscope.
## Summary of Results of Measurements on Höngschmid Standards

<table>
<thead>
<tr>
<th>Place</th>
<th>Method</th>
<th>Standards</th>
<th>Ratios</th>
<th>Höngschmid Ratios</th>
</tr>
</thead>
<tbody>
<tr>
<td>NBS</td>
<td>Electroscope</td>
<td>A/B</td>
<td>2.441</td>
<td>2.450</td>
</tr>
<tr>
<td>NBS</td>
<td>Calorimeter</td>
<td>A/B</td>
<td>2.450</td>
<td></td>
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<tr>
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<td>A/D</td>
<td>1.870</td>
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<tr>
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<td>A/D</td>
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<td>D/B</td>
<td>1.305</td>
<td>1.310</td>
</tr>
<tr>
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<td>Calorimeter</td>
<td>D/B</td>
<td>1.308</td>
<td></td>
</tr>
<tr>
<td>NBS</td>
<td>GM Counter</td>
<td>D/B</td>
<td>1.306</td>
<td></td>
</tr>
<tr>
<td>NPL</td>
<td>Electroscope</td>
<td>D/B</td>
<td>1.304</td>
<td></td>
</tr>
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<td>D/B</td>
<td>1.306</td>
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<tr>
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<td>GM Counter</td>
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<td>A/G</td>
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<td>D/G</td>
<td>1.398</td>
<td></td>
</tr>
</tbody>
</table>

### Energy dissipation from Po²¹⁰ as a function of time

<table>
<thead>
<tr>
<th>Date</th>
<th>Time (EST)</th>
<th>Hours from zero</th>
<th>Observed energy emission in microwatts</th>
<th>Energy emission corrected to zero date* (using half-life of 138.39 days) in microwatts</th>
</tr>
</thead>
<tbody>
<tr>
<td>22 June 1954</td>
<td>13.30</td>
<td>−167.75</td>
<td>1323.6</td>
<td>1278.1</td>
</tr>
<tr>
<td>29 June 1954</td>
<td>13.15</td>
<td>0</td>
<td>1276.9</td>
<td>1276.9</td>
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<tr>
<td>6 July 1954</td>
<td>15.05</td>
<td>169.75</td>
<td>1235.2</td>
<td>1279.8</td>
</tr>
<tr>
<td>13 July 1954</td>
<td>14.40</td>
<td>337.25</td>
<td>1190.5</td>
<td>1277.3</td>
</tr>
<tr>
<td>20 July 1954</td>
<td>14.20</td>
<td>505.25</td>
<td>1151.2</td>
<td>1279.2</td>
</tr>
<tr>
<td>24 August 1954</td>
<td>13.25</td>
<td>1344.25</td>
<td>961.43</td>
<td>1272.8</td>
</tr>
<tr>
<td>31 August 1954</td>
<td>10.05</td>
<td>1508.75</td>
<td>929.29</td>
<td>1273.2</td>
</tr>
<tr>
<td>7 September 1954</td>
<td>14.30</td>
<td>1681.25</td>
<td>899.37</td>
<td>1277.5</td>
</tr>
</tbody>
</table>

* Average value is 1276.9 microwatts with a standard deviation of 0.2%.
We made use of the microcalorimeter to determine the activity of a source of $\text{Po}^{210}$ in measuring the branching ratio in the decay of $\text{Po}^{210}$. We used about a 50 mc source of $\text{Po}^{210}$, and measured its activity by obtaining its rate of energy emission, using the energy of the $\alpha$ particles to give the disintegrations per second; a five-inch crystal was used to determine the $\gamma$-ray emission to get the branching ratio (figure 5).

I just want to run briefly through the rest of the various activities of our section at the National Bureau of Standards. Figure 6 shows our $\alpha$ standardization laboratory where we produce $\alpha$ particle sources on silver disks which are calibrated for $\alpha$ emission in a $2\pi$ geometry, irrespective of whether they are back-scattered or directly emitted $\alpha$ particles. Our $2\pi\alpha$ proportional counter has a diameter of about 5 inches, so as to obtain uniformity of response over the central region.

The workhorse of our section for the $\beta$ standards programme is of course the $4\pi\beta$ counter (figure 7) at it is in every laboratory, and a series of $4\pi\beta$ counters which we have used at various times are shown. Generally, however, we now use the one shown in figure 8, where the $4\pi\beta$ counter is open with the source in position. Figure 9 shows the various equipment we use for the preparation of the sources: micropipettes, ultramicroburettes and pycnometers.
The ultramicroburette shown consists of a steel plunger in mercury which forces out the liquid and you can deliver 0.1 ml with a precision of 1 in a 1000.

Figure 10 shows my colleague, Dr. R. W. Hayward, doing some measurements on Co$^{60}$ by coincidence counting, $\gamma$-$\gamma$ coincidence counting. A variable delay-line is used to vary the resolving time and then extrapolation of the coincidence counting rate to zero delay-line length is used to give the true coincidence counting rate without accidentals. The angular correlation between the directions of emission of the two $\gamma$-rays of Co$^{60}$ is used to obtain the disintegration rate under different experimental conditions in order to check for any spurious effects that may be present.

Figure 11 shows my colleague, Dr. H. H. Seliger, engaged in coincidence counting. This is a coincidence counting set-up with the $\beta$ counter on the right having a very thin, half-millimeter thick, crystal while the $\gamma$ counter on the left has a thicker crystal.

For low energy $\beta$ emitters such as C$^{14}$ and gaseous $\beta$ emitters such as Kr$^{85}$, a method of internal gas counting is used and figure 12 shows our gas counters out of their shield and out of their vacuum covers. On the left are three stainless-steel counters, one long and two short, one of which we call medium, however, and copper counters, one long and two short. I'll talk more about that tomorrow, if I may.
In our laboratories the radiochemical composition of our standard solutions is investigated with a view to achieving radiochemical stability. In figure 13 a sample of tritiated water has been broken under water in order to make the first dilution in the preparation of tritium standards. The tritiated water, which was about 4 curies in activity and which was contained in a quartz bulb under hydrogen to prevent dissociation, was measured in a microcalorimeter and was then crushed under a liter of water in a stainless-steel container. The 4 curies of tritiated water were diluted under gravimetric conditions and then further diluted to form our tritium standards. These tritium standards have been compared with those of the Chalk River Laboratories with an agreement of about 1—2%.

Figure 14 shows Mr. Marlow and Mr. Medlock preparing C\(^{14}\) standards in benzoic acid. These standards are for the calibration of liquid scintillation counters. The figure shows their being transferred into ampoules (containing about 3 ml of solution). They were then frozen while being flame-sealed.

For the relative measurement of β emitters or for the maintenance of reference standards of the short-lived β emitters we use a 2\(\pi\)β ionization chamber which is shown diagrammatically in figure 15. This is, in effect, a
standard instrument with which we maintain our standards and the constancy of the calibration of which is checked by means of a reference source of radium. The length of the collecting electrode here was adjusted quite empirically, so that a flat response was obtained when a point source was displaced slightly from the centre.

Figure 16 shows our $4 \pi \gamma$ chamber. This is also used in the same way as the $2 \pi \beta$ chamber for the preservation of the standardization of $\gamma$ emitters and
this has a quite flat response. The curves illustrate an experiment which has been carried out simply by taking one milliliter of Co\textsuperscript{60} solution in one of our ampoules and making a measurement, then adding 1 milliliter of carrier solution so that the same strength of source was in two milliliters and so on up to 5 ml. Even at 5 ml the chamber response is still about 99\% of that obtained with the 1 ml sample. The point is, however, that we only use 5 ml, so that we do not experience any such variations in volume. We use 5 ml of solution whenever we are carrying out our calibrations. This chamber quite fortuitously has a completely linear response from about 400 keV up to about 2 MeV and we can therefore use this to calibrate by interpolation a number of other $\gamma$ emitters. The precision of this chamber is 0.01\%. That is we can reproduce measurements on any source with this precision, but of course the accuracy is of the order of $\frac{1}{2}$\%. 

Figure 17 shows both our chambers in use. The manual controls shown use a Townsend-balance method with charge compensation, and the equipment on the right is an automatic device developed by Mr. Garfinkel for the same purpose. Three little photocells in the eyepiece of the Lindermann-Ryerson-electrometer telescope control relays to begin and end a run and to adjust the compensating voltage. The time of the run is measured by a decatron scaler operating on the standard Bureau of Standards kilocycle signal.
For relative measurements we also use a $2\pi\beta$ flow proportional counter, using organic solvents, such as formamide. Aqueous solutions are soluble in formamide and you can use up to about 10% aqueous solution without destroying the properties of your counter. We do not usually use quite as much as this. We dissolve the aqueous solution of the salt of the nuclide in question in formamide and put it in a carefully machined cell which is clamped up against the hemisphere of the counter. In the case of $\beta$ emitters greater than saturation thickness is used so that it does not matter how much you fill beyond a certain depth. This makes it even simpler. With $\gamma$ emitters one uses 1 ml of solution, the cell having been constructed to take 1 ml, and we can then obtain quite good comparative measurements (figure 18).

Figure 19 shows the finished standards being stored by M. Stockmann and Miss Cavallo until such time as they are required. The glass ampoules are packed with absorbent cotton inside a plastic container so that the solution will be absorbed in case of an accident.
I. 8. TECHNIQUES OF LARGE SCALE PROCESSING OF COUNTING DATA

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Abstract

The problems of a laboratory performing over 10,000 radionuclide analyses per year each of which requires a high degree of accuracy, but below that required in absolute standardization, call for special requirements in data handling and in data processing. Typical measurements involve mixtures of α emitters and multi-channel pulse-height analyses, 2π and 4π β counting, β-γ coincidence measurements.

Efficient use of instruments for measuring low levels of activity is achieved by the technique of multi-plexing with magnetic tape storage and read-out.

Arithmetical errors, particularly the troublesome ones involving inadvertent transposition of digits is virtually eliminated by the use of electronic calculators. In one typical operation involving a mixture of known nuclides, the gross counting data are fed into a machine which prints (reads-out) the answer on an electric typewriter. Alternatively, the results, if from a spectrometer, can be automatically plotted on an X-Y graph for further interpretive analysis. Block diagrams illustrating the sequence of steps are presented.

Decay corrections are easily handled by discarding the concept of the Gregorian calendar and employing instead a year in which the days are numbered consecutively beginning at midnight of the night of 31 December — 1 January, Greenwich Mean Time. For example, July 17, 1959, 3 : 5 PM Pacific Coast Daylight Saving Time is 198.920 days, and 2 : 30 PM October 16, 1959, Vienna time is 289.854. The advantages of being able to add, subtract, multiply or divide times expressed in this manner is apparent. The electronic circuit of a time clock based on this principle is described together with photographs.

Les techniques d’exploitation à grande échelle des données de comptage

Dans un laboratoire qui fait chaque année plus de 10,000 analyses de radionucléides, dont chacune doit avoir une grande précision, sans atteindre celle qui est requise aux fins de normalisation absolue, le dépouillement et l’exploitation des données posent des problèmes particuliers. On doit couramment procéder aux opérations suivantes: mesures de mélanges d’émetteurs α, analyses de hauteurs d’impulsions par multicanaux, comptage β 2π et 4π, mesures par coïncidences β-γ.

On assure une utilisation efficace des instruments de mesure de faibles niveaux d’activité en recourant à la technique “multiplex” avec enregistrement sur bande magnétique et dispositif de lecture.

Les erreurs de calcul, particulièrement les fautes d’inattention difficiles à retrouver, comme la permutation de deux chiffres, sont pratiquement
éliminées par l’emploi de calculatrices électroniques. Dans une opération courante portant sur un mélange de nuclides connus, les données brutes résultant du comptage sont communiquées à la calculatrice qui imprime la réponse finale au moyen d’une machine à écrire électrique. Si les résultats proviennent d’un spectromètre, on peut également en obtenir automatiquement une représentation graphique en coordonnées cartésiennes pour en faire l’analyse et en donner une interprétation plus poussée. Le mémoire contient des schémas d’opérations.

Pour opérer facilement les corrections de taux de désintégration, on remplace l’année du calendrier grégorien par une année dont les jours, sont numérotés consécutivement, et qui commence le 1er Janvier, à 0 heure de Greenwich. Par exemple, le 17 juillet 1959 à 15 h. 15 (heure d’été de la côte du Pacifique) sera le jour 198, 920 et le 16 octobre 1959 à 14 h. 30 (heure de Vienne) sera le jour 289, 854. Les avantages qui découlent de la possibilité d’ajouter, de retrancher, de multiplier et de diviser les temps exprimés de cette manière sont évidents. Le circuit électronique d’une horloge fondée sur ce principe est décrit dans le mémoire, qui contient en outre des photos.
Técnicas de tratamiento en gran escala de los datos obtenidos en el recuento

En un laboratorio donde se tengan que efectuar anualmente más de 10 000 análisis de radionúclidos con una precisión elevada, pero no tanto como la necesaria en los trabajos de calibración absoluta, el acopio, registro y tratamiento de los datos plantea problemas especiales. La metrología de los radionúclidos suele comprender las siguientes operaciones típicas: medición de la actividad de mezclas de emisores α, determinación de la amplitud de impulsos mediante analizadores de canales múltiples, recuento β y mediciones β-γ por el método de coincidencias.

El registro de datos en cintas magnéticas y la lectura de los mismos con arreglo al sistema "multiplex" asegura el aprovechamiento racional de los instrumentos utilizados en la medición de bajas actividades.

Los errores aritméticos, en especial las enojosas equivocaciones debidas a la transposición inadvertida de cifras, quedan prácticamente eliminados cuando se utilizan calculadoras electrónicas. En una operación corriente de medición de una mezcla de núclidos conocidos, los datos brutos obtenidos en el recuento se comunican a una calculadora que imprime el resultado mediante una máquina de escribir eléctrica. Si se trata de determinaciones espectrométricas, los resultados pueden representarse automáticamente en coordenadas cartesianas, lo que permite proceder a su análisis e interpretación. En la memoria se reproducen esquemas que indican el orden de las operaciones.

Las correcciones del tiempo de desintegración se facilitan sustituyendo el año del calendario gregoriano por un año en que los días están numerados consecutivamente, comenzado a medianoche del 31 de diciembre (hora del meridiano de Greenwich). Así las 15.05 horas del 17 de julio de 1959, hora de verano del Pacífico, se expresan, según este sistema, como día 198,920, y las 14.30 del 16 de octubre de 1959 hora centroeuropea), como día 289,854. Es evidente la ventaja de este método, que permite sumar, testar, multiplicar o dividir los tiempos expresados con arreglo al mismo. Se describe el circuito electrónico de un cronómetro basado en este principio; la descripción se ilustra con fotografías.

I Introduction

Recent advances in the arts of radiochemical analyses and in instrumentation have improved to the point where they can produce information faster than the human mind can absorb it and we are now turning to fast data-handling equipment to present the wealth of material in a more useful form.

The problems of a nuclear research and development laboratory performing a large number of radionuclide determinations per year call for special techniques in radiochemical analysis and in large-scale processing of the counting data.

In some cases, certified standards of radioactivity exist for particular nuclides, but in many others the laboratory is required to prepare its own working standards, to furnish them to other laboratories engaged in the same field of interest, and to compare the results of intercalibrations. These working
standards may be in the form of solutions of the radioactive nuclides including the rare earths or the transuranic elements.

Such laboratories in turn may use these working standards in their own research and development programs which may involve a study of activation analyses, start-up of a nuclear reactor, etc. Such practical programs may frequently involve the performance of many thousands of nuclide determinations per year.

The purpose of this paper is to present information concerning the manner in which such large numbers of radiochemical determinations and computations are performed and the techniques used to minimize human error and judgment. This information may be useful to laboratories contemplating expansion of their present programs for distribution of nuclide standards or of correlating the results of nuclide intercomparison. It may also be useful to those laboratories having responsibility for other functions in addition to the investigation of primary standards.

II Radiochemical Analyses

The analysis of a variety of samples possessing a wide range of activities and at the rate of several thousand measurements per year calls for careful organization and internal cross-checking and calibration. Such measurements are made possible by modifying the traditional concepts of chemistry and counting and by streamlining and automating the procedures wherever possible. No single individual performs all of the operations on any one sample, and the work is divided among specialists in the respective fields of chemical separations, counting, calculations and interpretation.

It is therefore important that a rigid system of "traffic control" be applied to the flow of samples and that the system be responsive to any accidents such as an occasional loss or inadvertent destruction of a sample.

Organization

The traffic control pattern for the analysis of samples, is shown in figure 1. After entering and logging the sample, and assigning it a code number indicating what isotopes are to be determined, the preliminary steps such as weighing, dissolution, addition of carriers, aliquoting, etc., are performed. The required chemical separations are next performed using precipitation, scavenging, ion-exchange, and other appropriate techniques. The samples are then mounted on planchets. The responsibility of the radiochemical analysis group ceases here.

Each planchet (or other prepared mounting) is then transferred to a separate group skilled in making the appropriate radioactivity measurements required of each planchet. Automatic sample changers are used quite liberally. Measurements of the $\alpha$, $\beta$, $\gamma$, or positron activity are made, as well as multichannel pulse-height analysis. The work of this group is extremely exacting and involves constant re-checking of the counting equipment.
Figure 1
Traffic control pattern for the analysis of samples.
The raw counting data are fed into an electronic computer and the finished data are presented as desired, for example, on punch cards, on electric typewriter, or an automatic X-Y plot as in the case of γ pulse-height analysis.

A fourth group interprets the data in terms of the program objective, scans the results for anomalies, and may request a re-run of a particular sample.

Quality Control

Quality control is maintained in several ways.

(1) A certain number of planchets after being counted and calculated are given a new code number and recycled into the counting room. The identities of these recycled samples are not apparent except to the Quality Control Group who compares the two sets of finished data and judges whether the degree of reproducibility of the counting (and calculations) operation is satisfactory in terms of the nature and the specific activity of the nuclide (dpm per gram) for which the analysis is being performed and the allowable errors (standard deviation) in counting.

(2) The National Bureau of Standards has issued standard samples of many radionuclides. After appropriate dilutions and aliquoting, these samples are used in two ways. In the first, known aliquots are evaporated onto appropriate planchets or other supporting materials, and the counting rate in terms of counts per minute less background is measured. The ratio of cpm/dpm yields the overall geometry factor for each radionuclide under the conditions of measurement. Daily variations in the value of the geometry factor are carefully examined and if necessary the suspected counting equipment is withdrawn from the operation line and the planchets are recounted.

The second use of the NBS standards is in the analysis of low levels of acticity or when a new chemical method of separation is being tested. For example, it may be known that short-lived Sr$^{85}$ (65 day) is absent from samples which are to be analyzed for low level activities of Sr$^{90}$. A known quantity of Sr$^{85}$ is added to the sample as a "spike". The chemistry of the two strontium isotopes is identical of course, and by counting the sample for Sr$^{85}$ and for Sr$^{90}$ and relating the Sr$^{85}$ count to the quantity originally added, it is possible to compute the radiochemical yield of the separation process. This correction factor compensates for losses of Sr$^{85}$ (and of Sr$^{90}$ in exactly the same ratio) because of adsorption on the walls of the beakers, centrifuge tubes, etc., and for losses due to finite solubility of precipitates.

(3) Low-level counting measurements pose special problems because of radiochemical impurities in the reagents used. The experience of this laboratory has made it mandatory to purify all reagents, including distilled water, that are to be used in low level counting. Traces of uranium have been found in HF and other reagents; not only does the U content vary, but the ratio of $^{235}$U/$^{238}$U as well.

Impurities are expressed colloquially in terms of "smidgins" or $10^{-9}$ grams of impurity. Table I lists some of the impurities found.
TABLE I
Radiochemical Impurities found in Reagents

<table>
<thead>
<tr>
<th></th>
<th>Uranium content, in Smidgins * per liter</th>
</tr>
</thead>
<tbody>
<tr>
<td>H₂O distilled, (average)</td>
<td>4</td>
</tr>
<tr>
<td>H₂O distilled twice from quartz</td>
<td>0.25</td>
</tr>
<tr>
<td>HNO₃, reagent grade</td>
<td>4.2</td>
</tr>
<tr>
<td>Fuming HNO₃, commercial</td>
<td>10</td>
</tr>
<tr>
<td>Fuming HNO₃, redistilled</td>
<td>5</td>
</tr>
<tr>
<td>HF commercial</td>
<td>10—15</td>
</tr>
<tr>
<td>HClO₄ reagent</td>
<td>5—10</td>
</tr>
<tr>
<td>HClO₄, distilled</td>
<td>2—5</td>
</tr>
</tbody>
</table>

* 1 smidgin = 10⁻⁹ g

Reagent blanks are determined as shown schematically in the right-hand portion of figure 1.

With this traffic and quality control in satisfactory operation, one gradually acquires confidence in the mass handling of samples.

A similar control procedure (not shown here) is applied to the analysis of samples by mass spectrographic methods.

III The Decimal Day Clock

Corrections for radioactive decay are generally made using conventional calendar time, expressed in terms of the appropriate month, hour, day, and minute. To compare the results of several measurements made in different parts of the world at different times is a process that is annoying and is subject to frequent errors, particularly if some of the locations are on Daylight Saving Time, and others are not.

The use of a decimal day clock starting at exactly midnight of the night of 31 December — 1 January, Greenwich Mean Time offers a great advantage compared to calendar time, because it is much easier to add, subtract, multiply or divide times by this method.

For example, 3:05 PM Pacific Coast Daylight Saving Time (USA) on July 17, 1959, is 198.920 days GMT and 4:30 PM, Vienna, Austria, time on October 14, 1959, is 285.729 days, GMT.

The construction of a decimal day clock is relatively simple. One version involves use of a synchronous motor operating at 1 rpm, actuated from the main line frequency (60 cps in the United States, 50 cps in Europe). Assume, for example, it is desired that the decimal day clock indicate visually the time to the nearest 0.001 day (86.40 seconds). For the 60 cycle motor, a gear with 25 teeth is fastened to the motor shaft and this gear drives another gear with 36 teeth. Because the synchronous motor operates at exactly 1 rpm, the second gear makes one revolution exactly every 86.4 seconds. (The accuracy of the clock will be as accurate as the average line frequency over the period of the measurements.)

Once per revolution, a cam fastened to the shaft of the second gear momentarily closes a microswitch and delivers a pulse to a 10-position
stepping switch. This switch advances one position every 86.40 seconds or 0.001 days. A second stepping switch is advanced one position when the first switch reaches its tenth position, and so on to as many stepping switches as desired. A total of six stepping switches permits reading the time directly to the nearest 0.001 day with the use of a bank of neon Nixie tubes.

Figure 2 illustrates the decimal day clock. The numerals are clearly visible at a distance of 50 feet. The master unit houses the 1 rpm motor and the six-10 position stepping switches, and can be used to operate any desired number of banks of Nixie tubes. Each bank can be mounted on the wall of the desired room.

Figure 2
The decimal day clock

Provision is also made for wiring the output of the decimal day clock to a data printer or to the input of a computer. Power requirements for the entire unit, with one bank of six Nixie tubes are 60 watts at 110 or 220 volts AC. A reset button warns the operator if a power interruption has occurred.

A more sophisticated version of the decimal day clock eliminates the dependence upon accurate power-line frequency maintenance and upon the absence of power interruptions. A crystal-controlled transistorized oscillator operating at 100 kc can be readily constructed which will maintain its frequency to ±0.005%. (The frequency calibration can be easily checked whenever desired against a subharmonic from the National Bureau of Standards radio station WWV.) The output frequency of the oscillator is suitably divided to bring it down to a sub-frequency capable of operating a synchronous motor which in turn operates a gear train and a microswitch as in the simpler version above. Interruption of the main 110 V (or 220 V) power supply is compensated for by "floating" the system with a storage battery. The latter operates only when the main power is interrupted, and is kept
in a fully-charged condition by means of a rectified, low-voltage current obtained from the mains.

The decimal day clock should prove to be of increasing value as the trend continues toward automation of counting rooms. Even for laboratories using manual computation methods, the decimal day clock is well worth acquiring.

IV Role of Computers in Data Reduction

Applicability

Electronic computers have been assigned a vital role in processing the large volume of data that is normally generated in the operation of a modern radiochemical laboratory, because simple and fairly complex measurements can both be handled with a high degree of accuracy, efficiency, and economy. As a result of these factors, the application of computers has extended into every phase of data reduction. This has led to increased interest in the development of automated nuclear counting facilities capable of expressing the data in a form (or language) acceptable to the computer input.

In data reduction, accuracy is the most important consideration. Accuracy is heightened by the elimination of human error in the transcription of data and in the operation of a desk calculator. However, human judgment is maintained by incorporating into the calculation procedure suitable criteria for scrutiny of the data. The efficiency of a computer is related to the number of operations it can perform per unit time. Economy is related to the quantity of operations and the cost per operation. Table II presents the number of “equivalent” multiplication operations per second, the cost per operation and approximate values for the initial cost or monthly rental for various computers, including a desk calculator.

<table>
<thead>
<tr>
<th>Machine</th>
<th>Operations/Sec</th>
<th>Cost/Operation ($/sec)</th>
<th>Monthly Rental-$/mo</th>
<th>Purchase Price $</th>
</tr>
</thead>
<tbody>
<tr>
<td>Desk Calculator</td>
<td>$2.5 \times 10^{-2}$</td>
<td>$3 \times 10^{-2}$</td>
<td>$6 \times 10^1$</td>
<td>$0.8 \times 10^8$</td>
</tr>
<tr>
<td>Bendix G-15</td>
<td>$0.6 \times 10^3$</td>
<td>$5 \times 10^{-5}$</td>
<td>$2 \times 10^3$</td>
<td>$7 \times 10^4$</td>
</tr>
<tr>
<td>IBM 650</td>
<td>$1 \times 10^2$</td>
<td>$6 \times 10^{-5}$</td>
<td>$4 \times 10^3$</td>
<td>$2 \times 10^5$</td>
</tr>
<tr>
<td>Remington-Rand Solid-State</td>
<td>$1 \times 10^3$</td>
<td>$1 \times 10^{-5}$</td>
<td>$7 \times 10^3$</td>
<td>$4 \times 10^5$</td>
</tr>
<tr>
<td>IBM 704 R. M. 1103 A</td>
<td>$6 \times 10^3$</td>
<td>$1 \times 10^{-5}$</td>
<td>$4 \times 10^4$</td>
<td>$2 \times 10^6$</td>
</tr>
</tbody>
</table>

It is not always economical to use a computer. In some simple cases, for instance, the time required to prepare the computer input data may exceed the time required to do the calculation on a desk calculator.

Too few radiochemists realize the capacities and capabilities of today’s computers. Through the use of an electronic computer, Compton distribu-
tions can be determined, \( \gamma \) spectra resolved, and photopeak areas integrated quickly.

Graphical resolution by hand of decay curves containing three or more components is tedious, time-consuming, and subject to errors. Multiple regression is the optimum mathematical method of resolving multi-component curves. A multiple regression calculation, involving for example thirty observations of the gross activity from a four-component system, requires approximately ten hours using a desk calculator compared with five minutes on an IBM-650 Computer, including the time required to store the program and data in the computer's memory. However, the degree of accuracy obtainable with the computer is even more important than its speed. A small error early in the hand calculation (very probable, considering the number of operations involved) would result in large errors in the final answers. Table III presents the results of a computer calculation on a four-component sample of known composition measured over a two-month period.

<table>
<thead>
<tr>
<th>Component</th>
<th>( T_{1/2} )</th>
<th>Multiple Regression</th>
<th>Known Concentration (dpm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zr(^{95})</td>
<td>65 days</td>
<td>14.51%</td>
<td>14.51 ± 0.38%</td>
</tr>
<tr>
<td>Nb(^{95})</td>
<td>84 hr; 35 days</td>
<td>0.48%</td>
<td>0.50 ± 0.07%</td>
</tr>
<tr>
<td>Th(^{231})</td>
<td>26 hr</td>
<td>44.00%</td>
<td>44.03 ± 0.66%</td>
</tr>
<tr>
<td>Th(^{234})</td>
<td>24 days</td>
<td>41.01%</td>
<td>40.96 ± 0.64%</td>
</tr>
</tbody>
</table>

The presence of an unknown fifth component may affect the results considerably. However, its presence may be detected by comparing the decay rate predicted by the regression equation with the observed decay rate. An estimate of its half-life may then be made and the multiple regression procedure repeated on a five-component decay curve.

Programming

An illustration of a common type of calculation quickly solved by a computer is the conversion of the observed counts per minute to specific activity; that is, disintegrations per minute per unit weight, at a desired reference time. The input data required for this calculation consists of counter number, number of counts observed, background, counting time, decay constant, end of count time, sample thickness, chemical yield, reference time, and sample weight. The standard deviations of the background and the chemical yield are also fed into the computer. The processing of this data is facilitated by recording it on a convenient form sheet. All of this information is hand-punched into IBM cards or magnetic tape.

Figure 3 is a block diagram of the computer program which represents the sequence of instructions for this routine calculation.
Block Diagramm Computer Program
For Converting Counts per Minute
To Disintegrations per Minute

START

Block 1
Read and Store
Raw Data

Block 2
Calculate:
CPM, \( \sigma_{CPM} \)

Block 3
Correct for
death-time loss

Block 4
Subtract
background

Block 5
"Table-look-up"
Determine Counting
Efficiency

Block 6
"Table-look-up"
Determine Standard
Factor

Block 10
Calculate:
Specific Activity, \( \sigma_{S.A.} \)

Block 7
Correct for
decay during
count

Block 8
Calculate:
DPM at the end
of the count

Block 9
Correct DPM to
reference time

Block 11
Read out results

END
Block 1: Read and store the input in the computer's memory.

Block 2: Calculate the counts per minute (CPM) and standard deviation, $\sigma_{\text{CPM}}$.

- Recorded counts = scalers \times scale factor
- $\text{CPM}_1 = \frac{\text{counts}}{\text{counting time}}$
- $\sigma_{\text{CPM}_1} = \sqrt{\frac{\text{counts}}{\text{counting time}}}$

Store $\text{CPM}_1$ and $\sigma_{\text{CPM}_1}$ — Go to Block 3.

Block 3: Correct for dead-time losses (time during which the counter is insensitive to counts).

$$\text{CPM}_2 = \frac{\text{CPM}_1}{1 - \text{CPM}_1 \tau}; \quad \text{CPM}_2 = \frac{\sigma_{\text{CPM}_1}}{(1 - \text{CPM}_1 \tau)^2}$$

Where $\text{CPM}_2$ is the observed counting rate corrected for coincidence losses, and $\tau$ is the dead-time constant in reciprocal counts per minute.

Store $\text{CPM}_2$ and $\sigma_{\text{CPM}_2}$ and go to Block 4.

Block 4: Subtract background.

$$\text{CPM}_3 = \text{CPM}_2 - \text{Background CPM}$$

$$\sigma_{\text{CPM}_3} = \sqrt{\sigma_{\text{CPM}_2}^2 + \sigma_{\text{BG}}^2}$$

Block 5: Computer performs a "table look-up" to determine the counting efficiency, $E$, for the sample thickness involved. A table of counting efficiency versus sample thickness is stored in the computer. The appropriate efficiency is obtained by interpolation.

Store $E$ and go to Block 6.

Block 6: Determine the standard factor, $G$, for the counter used. The standard factor is defined as the ratio of the sample counting efficiency ($E_s$) on the counter used, to the sample counting efficiency ($E_t$) on the counter used to prepare the table.

$$G = E_s / E_t$$

The computer scans a table, stored in its memory, of standard factors for various counters.

Store $G$, and go to Block 7.

Block 7: Correct for sample decay during counting period.

$$(\text{CPM}_4 \pm \sigma_{\text{CPM}_4}) = (\text{CPM}_3 \pm \sigma_{\text{CPM}_3}) \times f$$

$$f = \frac{\lambda t_c}{(1 - e^{-\lambda t_c})}$$

where $t_c$ is the sample counting time and $\lambda$ the decay constant.

Store $\text{CPM}_4$ and $\sigma_{\text{CPM}_4}$, and go to Block 8.

Block 8: The disintegrations per minute at the end of the count, DPM$_1$, are calculated from the stored data.

$$\text{DPM}_1 = \frac{\text{CPM}_4}{\text{Yield} \times E \times G}$$

$$\left(\sigma_{\text{DPM}_1}\right)^2 = \frac{1}{Z^2} \left[ \left(\sigma_{\text{CPM}_4}\right)^2 + \left(\sigma_{\text{yield}} \times \text{CPM}_4 \times E \times G\right)^2 \right]$$

where $Z = \text{Yield} \times E \times G$

Store $\text{DPM}_1$ and $\sigma_{\text{DPM}_1}$, and go to Block 9.
Block 9: Correct DPM\textsubscript{i} to the desired reference time.
\[(DPM_{R} \pm \sigma_{DPM_{R}}) = (DPM_{i} \pm \sigma_{DPM_{i}}) e^{-\lambda t}\]
where \(t\) = end of count time — reference time.
Store DPM\textsubscript{R} and \(\sigma_{DPM_{R}}\) and go to Block 10.

Block 10: Calculate the specific activity, S. A., of the sample.
\[(S.A. \pm \sigma_{S.A.}) = (DPM_{R} \pm \sigma_{DPM_{R}})/\text{sample weight}\]
Store S. A. and \(\sigma_{S.A.}\) and go to Block 11.

Block 11: Read out the specific activity at a reference zero time, and an identification number. This completes the operation. Proceed to Block 1 and read the next input-data card.

Approximately one-half minute is required to hand-punch the initial set of input-data cards. Once the program is stored, an infinite amount of data relating to this program may be processed. Less than one minute is needed to store this program in the computer's memory and the operations described above in Blocks 1 through 11 are completed at the rate of about 100 cards per minute, or one sample every 1.7 sec.

Economics
What has been described above is neither the simplest nor the most complex system that can be devised.
Personnel requirements are about one-third that of a non-automated system. Thus in addition to greater accuracy, the automated system is considerably more efficient and more economical than the non-automated. Although computers involve a large initial outlay, the cost, if the computer is used steadily, is recovered generally in two years or less because of the savings in manpower. However, if the computer is to be used only for a few minutes per day, rental of computer time from an installation is less expensive. The virtual freedom from arithmetical errors (including a simple transposition of digits) becomes a bonus factor.

Availability of Computer Programs
Computer programs applicable to radiochemical calculations are available from several sources. The Nuclear Codes Group, a division of the US Atomic Energy Commission, publishes a quarterly Newsletter reporting available programs. Computer manufacturers keep a library of program abstracts in various scientific fields. These abstracts are available at their computer installations or business offices.
As a last resort a radiochemist can write his own program with the aid of a programmer's manual. Some manufacturers offer classes in programming, and/or skilled assistance.

The value of the computer is fourfold: accuracy, efficiency, economy, and diversity of application. At the present time, almost every radiochemical laboratory in the country has access to computer facilities. The initiative is left to the chemist.

To summarize, it is possible to perform both the radiochemistry and the calculations on a large scale by the methods which have been described. One gradually acquires confidence in using this system of “traffic control” for the radiochemistry, the cross-checking and calibration, the computers for the data reduction and the specialized group for interpretation of the results. It would be difficult indeed to go back to the old system.

*
I. 9. THE ORGANIZATION OF INTERNATIONAL INTERCOMPARISONS OF RADIOACTIVITY STANDARDS, WITH SPECIAL REFERENCE TO SUCH MEASUREMENTS OF NBS STANDARDS

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NATIONAL BUREAU OF STANDARDS, RADIOACTIVITY SECTION
WASHINGTON 25 D.C., USA

Abstract

The need for, and organization of, international inter-comparisons of radioactivity standards will be discussed and the results of some recent measurements will be reviewed.

Organisation de comparaisons internationales d'étalons radioactifs, notamment en ce qui concerne la mesure d'étalons NBS

La nécessité et l'organisation de comparaisons internationales d'étalons radioactifs sont examinées dans ce mémoire qui passe en revue les résultats de certaines mesures récentes.

Организация международного сопоставления эталонов радиоактивности с уделением особого внимания измерениям эталонов, которые проводятся Национальным бюро стандартов (США)

Будет обсуждена потребность в международном сопоставлении эталонов радиоактивности и организация этого дела, а также будут рассмотрены результаты некоторых последних измерений.

Organización de comparaciones entre los patrones de radiactividad de los diferentes países, con especial referencia a la medición de los patrones del National Bureau of Standards

Les autores examinan la necesidad de comparar en un plano internacional los patrones de radioactividad y de organizar esas comparaciones; discuten los resultados de algunas mediciones recientes.

Comparative measurements are effective in determining the best procedures for the development and maintenance of standards and methods of standardization. Apart from those of radium radioactivity standards have been evolving since 1946. They are, however, of limited value because of the constantly changing intensity of the material itself with time. For this reason, the International Commission of Radiological Units and Measurements recommended that the various national laboratories which issue radioactivity standards should also frequently compare them. Only in this way can we raise and maintain confidence in work based on the standards which are available in different parts of the world.

At first, the organizations participating in intercomparison work were those from the three countries which were then producing or distributing standards of radioactivity, namely the National Research Council’s labor-
atories at Chalk River, the National Physical Laboratory at Teddington and the National Bureau of Standards in Washington, D. C. International intercomparisons are now, however, no longer confined to the traditional national standardizing laboratories, but also include organizations such as the Commissariat à l'Energie atomique, the Fondation Curie, the Max Planck Institute, the Laboratorio de Fisica Nuclear, Santiago, the Atomic Energy Research Establishment at Harwell and the US Atomic Energy Commission. This program is being enlarged continuously to encompass those laboratories interested in obtaining and maintaining good agreement between absolute measuring techniques for some of the more commonly used short-lived radio-isotopes. (Figure 1: Participating Laboratories.)

### Laboratories Participating in Comparisons of Standards and Methods of Measurement Used

<table>
<thead>
<tr>
<th>LABORATORY</th>
<th>LABORATORY ISSUING STANDARDS</th>
<th>METHODS USED IN COMPARATIVE MEASUREMENTS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atomic Energy of Canada Limited Chalk River (AECL)</td>
<td>NPL, NBS</td>
<td>$4\pi\beta\ (PC)$, $4\pi\beta\ (PC)\ -\gamma$</td>
</tr>
<tr>
<td>Atomic Energy Research Establishment Harwell (AERE)</td>
<td>NBS</td>
<td>$4\pi\beta\ (GM)$, $\beta\gamma\ (GM)$</td>
</tr>
<tr>
<td>Commissariat à l'Energie atomique (CEA)</td>
<td>NPL, NBS</td>
<td>$4\pi\beta\ (GM)$, $4\pi\beta\ (PC)$, $\beta\gamma$, DSA</td>
</tr>
<tr>
<td>Fondation Curie France</td>
<td>NBS</td>
<td>$4\pi\beta\ (PC)$, $4\pi\beta\ (PC)$ -scin. $\gamma$</td>
</tr>
<tr>
<td>Institut für angewandte Radioaktivität Leipzig (IAR)</td>
<td>NPL</td>
<td>$4\pi\beta\ (GM)$, $\beta\ (GM)\gamma\ (GM)$</td>
</tr>
<tr>
<td>Kernreaktor, Bau- und Betriebs-Gesellschaft</td>
<td>NBS</td>
<td>$4\pi\ (PC)$</td>
</tr>
<tr>
<td>Laboratorio de Fisica Nuclear Santiago</td>
<td>NBS</td>
<td>$\beta\gamma$ coin., $2\pi\beta\ FC$</td>
</tr>
<tr>
<td>National Bureau of Standards Washington (NBS)</td>
<td>AECL, NPL</td>
<td>$4\pi\beta\ (PC)$, $4\pi\gamma\ IC$, $4\pi\beta\ (PC)\ -\gamma$, $\gamma\gamma$</td>
</tr>
<tr>
<td>National Physical Laboratory Teddington (NPL)</td>
<td>AECL, CEA, NBS</td>
<td>$4\pi\beta\ (PC)$, $4\pi\beta\ (PC)\ -\gamma$, $\beta\gamma$, $4\pi\beta\ (SS)$, $4\pi\gamma\ IC$</td>
</tr>
<tr>
<td>National Physical Research Laboratory Pretoria (NPRL)</td>
<td>NPL, NBS</td>
<td>$4\pi\beta\ (PC)$, $4\pi\beta\ (PC)\ -\gamma$, $4\pi\beta\ (LS)$, $4\pi\beta\ (LS)\ -\gamma$</td>
</tr>
<tr>
<td>National Research Council Ottawa (NRC)</td>
<td>AECL, NBS</td>
<td>$4\pi\beta$ scin. $\gamma$ coin., $4\pi\beta$</td>
</tr>
<tr>
<td>Max Planck Institute Gottingen (MPI)</td>
<td>CEA, NPL</td>
<td>$4\pi\beta\ (GM)$, $4\pi\beta\ (PC)$, $\beta\gamma$</td>
</tr>
<tr>
<td>Physikalisch-Technische Bundesanstalt Braunschweig (PTB)</td>
<td>NPL, NBS</td>
<td>$4\pi\beta\ IC$, $4\pi\beta\ (PC)$, $4\pi\beta\ -\gamma$ coin., $4\pi\beta\ (GM)$</td>
</tr>
</tbody>
</table>

Figure 1
By distributing strong samples of short-lived material, such as 15-hour Na\(^{24}\), measurements can be made in the various countries at ensuing zero times. Standards of Au\(^{198}\), sent by the National Bureau of Standards, were measured simultaneously in parts of the world as distant from each other as Canada and the Union of South Africa. (Figure 2 [Au\(^{198}\); Figure 3 [I\(^{131}\)].)

<table>
<thead>
<tr>
<th>Laboratory</th>
<th>Method</th>
<th>1500 GMT, November 21</th>
<th>1500 GMT, November 24</th>
<th>1500 GMT, November 25</th>
</tr>
</thead>
<tbody>
<tr>
<td>National Bureau of Standards</td>
<td>4 π β PC</td>
<td>1.306</td>
<td>1.295</td>
<td>1.290</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.320</td>
<td>1.309</td>
<td>1.310</td>
</tr>
<tr>
<td></td>
<td>4 π γ IC</td>
<td>1.306</td>
<td>1.309</td>
<td>1.304</td>
</tr>
<tr>
<td>National Physical Laboratory</td>
<td>4 π β PC</td>
<td></td>
<td></td>
<td>1.287</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>1.302</td>
</tr>
<tr>
<td></td>
<td>4 π γ IC</td>
<td></td>
<td></td>
<td>1.310</td>
</tr>
<tr>
<td></td>
<td>B-γ IC</td>
<td></td>
<td></td>
<td>1.315</td>
</tr>
<tr>
<td>National Physical Research Laboratory</td>
<td>4 π β-γ coin.</td>
<td></td>
<td>1.328</td>
<td>1.319</td>
</tr>
<tr>
<td></td>
<td>4 π β-γ coin.</td>
<td></td>
<td>1.327</td>
<td>1.331</td>
</tr>
<tr>
<td></td>
<td>4 π liq. scin.</td>
<td></td>
<td>1.326</td>
<td>1.316</td>
</tr>
<tr>
<td>National Research Council</td>
<td>4 π β-γ coin.</td>
<td>1.341</td>
<td>1.330</td>
<td>1.346</td>
</tr>
<tr>
<td>Physikalisch-Technische Bundesanstalt</td>
<td>4 π γ IC</td>
<td>1.325</td>
<td>1.287</td>
<td>1.283</td>
</tr>
<tr>
<td></td>
<td>4 π β PC</td>
<td>1.301</td>
<td>1.281</td>
<td>1.282</td>
</tr>
<tr>
<td></td>
<td>4 π β-γ coin.</td>
<td>1.329</td>
<td>1.333</td>
<td></td>
</tr>
<tr>
<td></td>
<td>4 π β PC</td>
<td>1.297</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>4 π β-γ coin.</td>
<td>1.325</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Density of the solution = 0.997 g/ml at 26° C.

Figure 2

Most radionuclide standards are in the form of salts dissolved in suitable carrier solutions in flame-sealed glass ampoules. The volume range is between 1 and 5 milliliters of solution; the activity range is of the order of 1 to 10 microcuries.

Results of measurements by different organizations generally agree to within a spread of 3%, in some cases there is a spread of 5%, and only occasionally are there wider differences. The spread does not seem to be a function of the number of laboratories participating. (Figure 4: International Comparison of Standards.)
International Comparisons of Standards

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Date</th>
<th>Laboratory Issuing Standard</th>
<th>Participating Laboratories</th>
<th>Spread in Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>K(^{42})</td>
<td>1958</td>
<td>NBS</td>
<td>AECL, NPL, NBS</td>
<td>±7%</td>
</tr>
<tr>
<td>P(^{32})</td>
<td>1958</td>
<td>AECL</td>
<td>NBS, NPRL, NPL, MBH, NRC, AECL</td>
<td>±7%</td>
</tr>
<tr>
<td>I(^{131})</td>
<td>1958</td>
<td>NPL</td>
<td>AECL, NBS, NPL, PTB, MPI</td>
<td>±3%</td>
</tr>
<tr>
<td>Na(^{24})</td>
<td>1958</td>
<td>NPL</td>
<td>AECL, NBS, NPL, PTB, IAR</td>
<td>±3%</td>
</tr>
<tr>
<td>Au(^{198})</td>
<td>1958</td>
<td>NBS</td>
<td>AECL, CEA, FC, MBH, LFN, NBS, NPL, NPRL, NRC, PTB</td>
<td>±5%</td>
</tr>
<tr>
<td>Na(^{22})</td>
<td>1959</td>
<td>NBS</td>
<td>AECL, FC, NBS, NPL</td>
<td>±5%</td>
</tr>
</tbody>
</table>

Figure 3

Summary of April 1958 I\(^{131}\) Intercomparison
Data Reduced to µc/g at 1500 GMT April 17, 1958

<table>
<thead>
<tr>
<th>Laboratory</th>
<th>Method</th>
<th>1500 GMT April 16, 1958</th>
<th>1500 GMT April 17, 1958</th>
<th>1500 GMT April 18, 1958</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atomic Energy of Canada Limited</td>
<td>4 (\pi \beta-\gamma) coin.</td>
<td>22.76</td>
<td>22.78</td>
<td>22.74</td>
</tr>
<tr>
<td>National Bureau of Standards</td>
<td>4 (\pi \gamma) IC</td>
<td>22.57</td>
<td>22.54</td>
<td>22.49</td>
</tr>
<tr>
<td></td>
<td>4 (\pi \beta) PC</td>
<td>22.56</td>
<td>22.55</td>
<td>22.73</td>
</tr>
<tr>
<td>National Physical Laboratory</td>
<td>Graphite IC</td>
<td>22.39</td>
<td>22.30</td>
<td>22.39</td>
</tr>
<tr>
<td></td>
<td>4 (\pi \beta) PC</td>
<td>22.31</td>
<td>22.36</td>
<td>22.39</td>
</tr>
<tr>
<td>Physikalisch-Technische Bundesanstalt</td>
<td>4 (\pi) GM</td>
<td>22.26</td>
<td>22.32</td>
<td>22.28</td>
</tr>
<tr>
<td>Max Planck Institute</td>
<td>4 (\pi) PC</td>
<td>22.17</td>
<td>22.20</td>
<td></td>
</tr>
</tbody>
</table>

Figure 4

In order to realize an objective of 1 or 2% accuracy for primary radioactivity standards, frequent intercomparisons between standardizing laboratories are needed.

Almost every nuclide requires its own method of standardization or some variation of an existing method which can be adapted to the kind and
energy of the radiations emitted, the chemical composition and reactions of the solution. It should be the responsibility of national laboratories to introduce their own technical procedures for the development and maintenance of standards. Information, such as that on the next figure, might prove useful, for along with participation, knowledge of work patterns and results provide guides for different and perhaps more refined techniques and, therefore, better standards. (Figure 5: International Intercomparison of Radioactivity Standards — NBS Na₂².)

National Bureau of Standards
Washington 25, D C
International Intercomparison of Radioactivity Standards
NBS Na₂²
Reporting Laboratory: National Bureau of Standards
To 1500 GMT February 25, 1959

VALUES

<table>
<thead>
<tr>
<th>Method</th>
<th>Dilution Factor</th>
<th>No. of Sources</th>
<th>Source Mount</th>
<th>Source Treatment</th>
<th>Activity of Ampoule Solution</th>
</tr>
</thead>
<tbody>
<tr>
<td>4πβPC</td>
<td>12.543</td>
<td>6</td>
<td>Au evap. collodion</td>
<td>Evap. in air</td>
<td>3.901 ± 4% × 10⁴</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Al leaf on collodion</td>
<td></td>
<td>3.919 ± 0.8%</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Al leaf on F-P</td>
<td></td>
<td>3.867</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Al leaf on F-P</td>
<td>2 drops 1/50 dilution of Tween evap. in air</td>
<td>3.935 ± 0.5%</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Al leaf on F-P</td>
<td>2 drops 10⁴ dilution ludox evap. in air</td>
<td>3.921 ± 0.3%</td>
</tr>
<tr>
<td></td>
<td>18.323</td>
<td>9</td>
<td>Al leaf on F-P</td>
<td>Freeze-dried</td>
<td>3.811 ± 0.7%</td>
</tr>
<tr>
<td></td>
<td>21.325</td>
<td>7</td>
<td>Al leaf on F-P</td>
<td>1 drop 10⁴ ludox evap. in air</td>
<td>3.917 ± 0.4%</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Al leaf on F-P</td>
<td>Evap. in air</td>
<td>3.776 ± 0.2%</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Au evap. F+P</td>
<td>1 drop 10⁴ ludox</td>
<td>3.918 ± 0.4%</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Au evap. collodion</td>
<td>2 drops and 4 drops 2 × 10⁴ ludox</td>
<td>3.989 ± 0.5%</td>
</tr>
<tr>
<td>4πγIC</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>4.014 ± 0.3%</td>
</tr>
<tr>
<td>4πβ-γ Coinc.</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>4.499 ± 0.3%</td>
</tr>
</tbody>
</table>

Figure 5
## CORRECTIONS

<table>
<thead>
<tr>
<th>Method</th>
<th>Background</th>
<th>Dead Time</th>
<th>Film Abs.</th>
<th>Decay</th>
<th>Half-life Used</th>
<th>Other</th>
</tr>
</thead>
<tbody>
<tr>
<td>4πβ PC</td>
<td>~ 1% Cr. rate</td>
<td>&gt; 0.1%</td>
<td>1—2%</td>
<td>0.1—9%</td>
<td>2.576 yrs.</td>
<td></td>
</tr>
<tr>
<td>4πγ IC</td>
<td>~ 1%</td>
<td></td>
<td></td>
<td>~ 3%</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
| 4πβ-γ Coinc. | γ Bkgd.   | ~ 14%     |           | ~ 10%  |                | Accidental coincidences = 0.4% | EC $\beta^+$ = 0.111
I. 10. STANDARDIZATION OF RADIONUCLIDES AT THE ATOMIC ENERGY COMPANY (SWEDEN)

JOSEF BRAUN

ATOMIC ENERGY COMPANY, SWEDEN

Abstract

The equipment in use at our laboratory for absolute measurement is described.

A survey is given over the results of β standardization performed with 4π and 2π proportional counters. An apparatus used for absolute measurement of gaseous isotopes is described.

Introduction

The development of the peaceful uses of atomic energy in Sweden has been concentrated to the partly state owned company, AB Atomenergi. As it was obvious already at an early stage that the programme would also include production of isotopes, a group for absolute measurements has been formed within the section for General Physics.

The first task has been to procure and investigate suitable equipment for our laboratory for absolute measurement, to develop appropriate measure-
ment techniques as well as testing methods by comparing certain results with measurements performed by NPL, AERE and NBS. The group is also responsible for a number of standardizations for other Swedish research institutions. This stage is now to be considered as finished. In connexion with the start of our new research reactor R2, type MTR, during the next year, the laboratory will move to the company's new research station, Studsvik, 70 miles south of Stockholm, in order to handle routine standardization of isotopes produced in the reactor.

Figure 1

1 Proportional counter for $4\pi$-geometry
2 Dismantled electrode system
3 Lower part to be used in $2\pi$-geometry
4 Window to be used in connexion with external counting

Our basic tool for the standardization of $\beta$ emitters is a $4\pi$ proportional flow-counter (figure 1). It consists of two $2\pi$ units, joined together by a spacing piece, in which runs the test slide. Sealing against the slide is made with O-seals. By replacing the lower part with a blank piece, the chamber can also be used for internal $2\pi$ counting. Special windows are available to be used for conventional external counting.
Figure 2 shows an intersection of the counter. The body is made of stainless steel with a thickness of 1 cm. The surfaces are treated with a mixture of acids, water and a wetting agent. The anode-connector has an isolation of Teflon. The loop is made of 0.03 mm or in some cases of 0.05 mm tungsten wire, which has been electropolished in a 20% NaOH solution, with 6.3 V AC and a current density of 6 A/cm². The spacing piece, gas nipples and other minor details are made of chromium plated brass.

![Figure 3](image)

**Figure 3**

High-voltage characteristics for isotopes with various maximum β energies.
The high voltage characteristics for various β emitters are shown in figure 3. The slope was between 0 and 0.3% per 100 V. The curves are obtained with a 0.03 mm loop. A 0.05 mm loop shifted the plateaus about 500 V to the right. With an over-voltage of 200—300 V the slope of the bias was insignificant. Figure 4 shows the influence of the amplifiers non-overloading characteristics on the counting rate. The comparison is done between a TMC amplifier type A 1-4 A and a very high class amplifier of type DD 2. In our β counting equipment the upper limit for negligible dead-time losses due to the amplifier seems to be about 6000 cps with delay-line clipping.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Amplifier</th>
<th>A 1-4 A difference in % of DD 2 value</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>DD 2</td>
<td>2 µs</td>
</tr>
<tr>
<td>Sr90—Y90</td>
<td></td>
<td>—</td>
</tr>
<tr>
<td>904</td>
<td></td>
<td>0.4</td>
</tr>
<tr>
<td>2535</td>
<td></td>
<td>0.3</td>
</tr>
<tr>
<td>6141</td>
<td></td>
<td>1.1</td>
</tr>
<tr>
<td>20985</td>
<td></td>
<td>4.3</td>
</tr>
<tr>
<td>Po214</td>
<td>835</td>
<td>—</td>
</tr>
<tr>
<td>2705</td>
<td></td>
<td>0.1</td>
</tr>
<tr>
<td>15830</td>
<td></td>
<td>1.8</td>
</tr>
<tr>
<td>37750</td>
<td></td>
<td>—</td>
</tr>
</tbody>
</table>

Figure 4
Difference in counting rate between A 1 and DD 2 for various clipping times and for delay-line clipping.

Figure 5
Gas amplification, in methan, lat
Figure 5 shows the gas amplification measured with methane (99% pure). The curves fit well to the theoretical expression given by Segré. Measurements of the pulse form have shown a rise-time below 0.1 μs.

4π measurements

As source mounts we use either aluminium foils of about 0.8 μ (200 μg/cm²) thickness or plastic foils. We tried both the combination Formvar reinforced with Zapon for chemical protection and VYNS. A normal value of surface density is 20 μg/cm² for Formvar & Zapon and 10 μg/cm² for VYNS. The conducting layer on the surface of the plastic films consists of evaporated gold of about 7 μg/cm² thickness, which may be compared with 2 μg/cm² the saturation thickness given by Pate and Yaffe.

Corrections

Counter efficiency is considered to be 100%, which is concluded from measurements with varying diameter of source and source mounting. Dead-time corrections are increasing linearly from 0.2% at 200 cps to 1% at 700 cps. (Valid for 5 μs clipping time, and may be reduced for delay-line clipping.) The correction may be regarded as accurate within 0.1%.
Source-mount absorption is shown in figure 6. It is apparent that a linear extrapolation back to 0 from Al-foil thickness gives a correction which is too low. The error in the extrapolation is estimated to 0.1—0.3%. Self-absorption has been compared for Al-foils and plastic foils both treated and untreated with a wetting agent. In all cases the results are in favour of Al-foils. Figure 7

<table>
<thead>
<tr>
<th>Source preparation</th>
<th>Ca$^{45}$ (0.25 MeV)</th>
<th>S$^{35}$ (0.167 MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>direct evaporation</td>
<td>direct evaporation</td>
</tr>
<tr>
<td>plastic films</td>
<td>32.5</td>
<td>20—43</td>
</tr>
<tr>
<td></td>
<td>insulin pretreatment</td>
<td>Tween pretreatment</td>
</tr>
<tr>
<td></td>
<td>34.6</td>
<td>34</td>
</tr>
<tr>
<td></td>
<td>oxalate precipitate</td>
<td>insulin pretreatment</td>
</tr>
<tr>
<td></td>
<td>34.6</td>
<td>42.4</td>
</tr>
<tr>
<td>sources on Al foils, directly evaporated</td>
<td>37.0</td>
<td>sources on Al foils, directly evaporated</td>
</tr>
<tr>
<td>from solution and corr. foil absorption</td>
<td></td>
<td>from solution and corr. foil absorption</td>
</tr>
<tr>
<td></td>
<td>48.3</td>
<td>48.3</td>
</tr>
</tbody>
</table>

Figure 7
Influence of source-preparation on self-absorption.

gives results for Ca$^{45}$ and S$^{35}$ obtained with different source preparations. The error of self-absorptions is estimated to 0—1% for $E_{\text{max}} > 0.5$ MeV and 2—3% for Ca$^{45}$ and S$^{35}$. The accuracy for Co is a poor one mainly on account of the thickness of the deposits. Figure 8 shows a comparison between our results and measurements performed at NBS and Nuclear Chicago Corporation in USA and NFL and Harwell in England.

2 $\pi$ counting

By using appropriate backing material and by careful source preparation, back-scattering coefficients may be measured with good accuracy and reproducibility. Therefore it is possible to employ 2 $\pi$ counting in cases where high precision is not so important. Figure 9 gives the result of measurement of back-scattering for 5 energies and 3 different materials. To improve the efficiency in the outer part of the counter a cylindrical screen was introduced around the loop holder. The screen terminated in a couple of plane lobs in parallel to the plane of the loop. With this arrangement (as shown in figure 2) a remarkable improvement was achieved in the high voltage characteristics of the counter. This is shown in figure 10. A sample of Y$^{91}$ (1.55 MeV) with a diameter of 30 mm (counter diameter = 40 mm) could now be measured with only about 0.5% lower efficiency compared with a centered point source. It was estimated that 2 $\pi$ standardization for $\beta$ emitters above 0.5 MeV can be performed on a routine base with an accuracy of about 3—5%.
**DISCUSSION**

**Professor Aten** (Netherlands):

I should like to ask Mr. Robinson whether any data are available on the efficiency at different neutron energies of the long counter mentioned in his paper. If a reliably calibrated standard long counter were available, the calibration of neutron sources would be enormously simplified.

**Dr. B. M. W. Robinson** (United Kingdom):

I am sorry, but we have no data so far on the efficiency at various neutron energies. Of course we realize the importance of this matter and intend to tackle it as soon as we can.

**Professor Aten**:

I should like to have the views of Mr. Grinberg and Mr. Robinson on the cost of the electric circuit used in conjunction with the standard ionization chamber. As far as the inter-comparison of \( \gamma \)-emitting samples is concerned, it might be asked whether equivalent results could not be obtained by using a \( 4 \pi \gamma \) Geiger-Müller counter.

**Dr. Grinberg** (France) (translated from French):

The electronic equipment required for the \( 4 \pi \gamma \) chamber is simply a DC amplifier which costs about 200,000 francs. I do not think that a Geiger-Müller \( 4 \pi \) counter can in any way replace the \( 4 \pi \gamma \) chamber; in any case, the electronic equipment required for the \( 4 \pi \) Geiger-Müller counter would be more expensive than that used with the ionization chamber.

**Professor Aten**:

The price of a simple counting set to go with a \( 4 \pi \gamma \) counter is roughly the same as that quoted by Dr. Grinberg. The price of the counter itself is negligible compared to the price of the electronics system, and I do think that Geiger-Müller counters have gone out of fashion in a way which is not quite deserved. Hand in hand with the improvement of other measuring instruments has gone the improvement of industrially available Geiger-Müller counters; we have had different types of Geiger-Müller counters for the preservation of standards of short-lived isotopes which have kept stable within 1 or 2% for periods of years. I should like to add that the \( 4 \pi \gamma \) counter has the same advantage as \( 4 \pi \gamma \) ionization chamber: that it is quite insensitive to the exact location of the sample.

**The Chairman** (Prof. O. Kofoed-Hansen, Denmark):

Thank you, Professor Aten, for your statement regarding the Geiger-Müller counters. I would now ask Dr. B. M. W. Robinson if he would care to answer Professor Aten's question regarding the cost of the electric equipment going with his standard ionization chamber.
<table>
<thead>
<tr>
<th>Isotope</th>
<th>$\beta$ max. energy MeV</th>
<th>Aluminium</th>
<th>Stainless</th>
<th>Platina</th>
<th>corr.*) for stainless</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ca$^{40}$</td>
<td>0.25</td>
<td>—</td>
<td>0.373 ± 0.002</td>
<td>—</td>
<td>0.373</td>
</tr>
<tr>
<td>I$^{131}$</td>
<td>0.60 (mean value)</td>
<td>—</td>
<td>0.500 ± 0.006</td>
<td>—</td>
<td>0.476</td>
</tr>
<tr>
<td>Tl$^{204}$</td>
<td>0.77</td>
<td>0.384 ± 0.001</td>
<td>0.482 ± 0.008</td>
<td>0.61 ± 0.01</td>
<td>0.482</td>
</tr>
<tr>
<td>Au$^{198}$</td>
<td>0.96</td>
<td>—</td>
<td>0.497 ± 0.010</td>
<td>0.67 ± 0.01</td>
<td>0.479</td>
</tr>
<tr>
<td>Y$^{91}$</td>
<td>1.55</td>
<td>—</td>
<td>0.482 ± 0.010</td>
<td>—</td>
<td>0.482</td>
</tr>
</tbody>
</table>

*) corrected for internal conversion.

Figure 9

Figure 10
The end effect in a counter of this type comes partly from disturbances at the boundaries of the sensitive volume and partly from β particles entering this volume from outer parts of the tube. By using a pair of tubes equal to each other in every aspect except the sensitive length the end effect can be eliminated.

The wall effect is due to β particles which are born near the wall and which may leave the counter without ionizing the gas. It can be shown that in a cylindrical counter this effect reduces the sensitive volume by a factor

\[
1 - \frac{1}{SD} - \frac{1}{K}
\]

where
- D diameter of tube in cm
- S specific ionization of β particle in the gas (ion pairs/cm)
- K probability for a β particle to produce a secondary electron in the wall which is back-scattered into the counter.

Measurements of the two correction terms mentioned above have been made with an arrangement of four tubes with two different lengths and two different diameters, giving the combination long—thin, long—thick, short—thin, short—thick.

The agreement between theoretical and measured wall corrections obtained with Kr\textsuperscript{85} (E\textsubscript{max} 740 keV) is good. Some values are given in the table below.

<table>
<thead>
<tr>
<th>L × D mm</th>
<th>End effect</th>
<th>Wall effect</th>
<th>Wall effect (calculated)</th>
</tr>
</thead>
<tbody>
<tr>
<td>71.7 × 24.7</td>
<td>12.0 ± 2.0</td>
<td>5.0 ± 2</td>
<td>4.5 ± 1</td>
</tr>
<tr>
<td>141.0 × 24.7</td>
<td>6.2 ± 1.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>279.0 × 24.7</td>
<td>3.0 ± 0.5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>92.0 × 31.8</td>
<td>7.2 ± 1.0</td>
<td>3.7 ± 1</td>
<td>3.5 ± 0.7</td>
</tr>
<tr>
<td>195.0 × 31.8</td>
<td>3.5 ± 0.5</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The filling system of the equipment is shown in figure 12.
Professor Aglintsev (Translated from Russian):

In the case of a number of radioisotopes, the Institute of Metrology has been carrying on research on their half-lives. In those instances where we have had to give a half-life of an isotope determined by us at the Institute, we have been giving the value found by us but, of course, taking into account the data in the literature; i.e. we have been giving the average of our own determinations and the published data. In instances where some isotope is concerned whose half-life has not been determined at the Institute, we have naturally been limiting ourselves to giving the most reliable published data. In order that the user can evaluate the data, we indicate the source from which they have been obtained.

Dr. Grinberg (Translated from French):

I have an important question to put, not only to Professor Aglintsev, but also to Dr. B. M. W. Robinson and Dr. Mann. It concerns the problem of radiochemical impurities contained in radioactive solutions, which Professor Aglintsev has said do not exceed 0.5%. I should like to know whether, and if so how, the nature and percentage of such radiochemical impurities is established and also whether such research work is carried on systematically in the various laboratories.

Professor Aglintsev (Translated from Russian):

Three methods are used to determine the degree of purity of a preparation. The first one is somewhat lengthy and is based on the half-life; the second is considerably more rapid and is based on the $\beta$ or $\gamma$ spectrum; and the third is one of several methods based on analysis of absorption curves and exploiting the fact that a pure preparation will give a single absorption rate while radiochemical admixtures will alter that rate. By combining these three methods we can arrive at some idea of radiochemical purity. I may perhaps add here that radiochemical purity is partially checked before the given preparation is readied by a number of methods, for example, by activation analysis or by careful refinement of the source materials from which the isotopes are prepared.

Dr. Mann (United States of America):

I am afraid that the methods of checking radiochemical purity at the National Bureau of Standards are rather simple ones, because we have rather a big demand for standards and not very many people to meet this demand. So radiochemical purity — over which we have no control, because we get the samples from Oak Ridge — is usually checked in the case of short-lived radionuclides by following the half-life. The other question with which we are concerned is that of radiochemical stability. This is usually checked by opening standards at various intervals under the conditions under which they would be used in the case of $\beta$-emitters, which are the ones with which we are chiefly concerned because the $\gamma$-emitting standards are usually used sealed. A case in point is $\text{Tl}^{204}$, where, by opening standards over a period of years and measuring the half-life, we obtain the value of 2.7 years which is often given in the literature, whereas with identical material which had been put into the form of a solid source at the beginning of its period, we
<table>
<thead>
<tr>
<th>Isotope</th>
<th>Supplied by</th>
<th>Supplier's</th>
<th>Method of standardisation</th>
<th>Stated disintegration rate Arbitrary units</th>
<th>Our value of the disintegration rate Arbitrary units</th>
</tr>
</thead>
<tbody>
<tr>
<td>P32</td>
<td>NBS</td>
<td>11.7.57</td>
<td>β ion chamber 1)</td>
<td>250 ± 2%</td>
<td>252</td>
</tr>
<tr>
<td>P32</td>
<td>NCC</td>
<td>9.9.57</td>
<td>β ion chamber 2)</td>
<td>250 ± 3%</td>
<td>262 ± 2%</td>
</tr>
<tr>
<td>P32</td>
<td>NPL</td>
<td>18.12.57</td>
<td>4π</td>
<td>250 ± 3%</td>
<td>255x 254</td>
</tr>
<tr>
<td>Na24</td>
<td>NPL</td>
<td>25.9.57</td>
<td>β counter</td>
<td>250 ± 3%</td>
<td>251</td>
</tr>
<tr>
<td>Na24</td>
<td>NCC</td>
<td>15.10.57</td>
<td>4π ion chamber 4)</td>
<td>250 ± 3%</td>
<td>244</td>
</tr>
<tr>
<td>Au198</td>
<td>NPL</td>
<td>20.11.57</td>
<td>β counter</td>
<td>250 ± 3%</td>
<td>252x 250 ± 2%</td>
</tr>
<tr>
<td>Tl204</td>
<td>AERE</td>
<td>12.12.57</td>
<td>4π</td>
<td>250.0 ± 3%</td>
<td>244 250 ± 3%</td>
</tr>
<tr>
<td>Sr90—Y90</td>
<td>NPL</td>
<td>3.6.57</td>
<td>β counter</td>
<td>250 ± 3%</td>
<td>247 250 ± 2%</td>
</tr>
<tr>
<td>I131</td>
<td>NPL</td>
<td>23.10.57</td>
<td>γ ion chamber 7)</td>
<td>250 ± 5%</td>
<td>250x 253 ± 3%</td>
</tr>
<tr>
<td>Co60</td>
<td>AERE</td>
<td>10.12.57</td>
<td>β-γ coincidence</td>
<td>250.0 ± 2%</td>
<td>228x 232x ± 10%</td>
</tr>
<tr>
<td>Ca45</td>
<td>AERE</td>
<td>12.12.57</td>
<td>4π</td>
<td>250.0 ± 3%</td>
<td>254x 250 ± 4%</td>
</tr>
<tr>
<td>S85</td>
<td>AERE</td>
<td>13.12.57</td>
<td>β counter</td>
<td>250.0 + 4% − 2%</td>
<td>249x 248x ± 4% − 2%</td>
</tr>
<tr>
<td>Ru106—Rh106</td>
<td>AERE</td>
<td>11.12.57</td>
<td>4π</td>
<td>250 ± 4% − 2%</td>
<td>251x 10 ± 3%</td>
</tr>
</tbody>
</table>

1)—10) See report AE-5.

Figure 8

Internal gas counting

For absolute measurement of inert gases and low energy β emitters, which are convertible into a gaseous state, we use the method of internal gas counting with GM tubes. The filling consists of a mixture of a known amount of the active-gas sample and a counting gas [10% CH3(OCH3)2, trade-name methylal + 90% argon at 140 mm Hg].

A typical GM tube is shown in figure 11. It is made of pyrex glass and has a transparent semiconducting cathode consisting of a tin compound. In order to obtain a fixed sensitive length the anode wire is shielded at both ends with a thin steel tube (O. D. 0.7 mm).
Dr. B. M. W. Robinson:

I am sorry, I do not know the exact cost, but Mr. Putman and I think £150 would be not far from the mark. That is for the commercially available AVO DC amplifier suitable for use with the NPL β-γ reference ionization chamber, but of course any laboratory can choose its own way of measuring current and very often one can find equipment which is not so expensive.

The Chairman:

I have a question here from Dr. Yaffe (Canada) addressed to Professor Aglintsev (USSR). Dr. Yaffe please.

Dr. Yaffe (Canada):

I would like to ask Professor Aglintsev if he would be good enough to describe the 4π counter which he mentioned very briefly in his paper on methods of standardization. I would be particularly interested in the counter itself, physically, the methods of source preparation and the various correction factors which are used. The reason quite frankly is that I have seen nothing of this in the literature and I am certain that the USSR has a great deal to contribute.

Professor Aglintsev (Union of Soviet Socialist Republics) (Translated from Russian):

The 4π counters we use have been described in the literature since 1956 and are of fairly ordinary design. We use counters consisting of 2 half-cylinders and 2 hemispheres. The preparation is applied in the usual way in a thin film and the general procedure has no special features. As to the question of accuracy, we are able to obtain very high accuracy on the basis of statistics. It is self-evident that 4π counters cannot be used to measure preparations measured in liquid scintillation counters.

Dr. Yaffe:

I appreciate the problem of statistics, and this is very easily solved, I agree, by length of duration of count. But the problems that interest me specifically are those concerning methods of correction for absorption in the source mount. For example, on what type of material are these sources mounted? The problem of self-absorption is a very serious one — how are the corrections for this made?

Professor Aglintsev (Translated from Russian):

A usual source whose diameter is normally 10—12 mm is mounted on an acetate film. The corrections are introduced by normal extrapolation methods. If the source is a weightless one, such corrections in practice cannot be made. The pulses in each half are calculated separately and the coincidences are calculated. The methods of 4π counting used in the USSR do not differ from those generally employed.
Dr. Grinberg (Translated from French):

I have several questions for Professor Aglintsev.

My first question is that I should like to know what techniques are used for measuring high-activity standard alpha sources. Are they measured with the continuous ionization chamber?

The second question also concerns alpha sources. Professor Aglintsev gave some details on the thickness of an electrolytically deposited source and I should like to know how these thicknesses are determined.

Professor Aglintsev (Translated from Russian):

Alpha sources are measured in a fast-acting, electron-collecting argon-filled ionization chamber. Generally speaking the form of such chambers differ. In some cases lattice chambers are used. That answers the first question.

Comparatively weak sources are measured by 2 or 4 \( \pi \) geometry; strong sources are measured at low solid-angle values.

Perhaps Professor Bochkarev could reply to the second question in relation to plutonium sources.

Professor Bochkarev (Union of Soviet Socialist Republics) (Translated from Russian):

The thickness of such sources is, as a rule, not checked for each individual sample, but it is calibrated by the strength of the current and selectively controlled by weighing.

Dr. Grinberg (Translated from French):

My third question relates to solid \( \beta \)-emitting calibrated sources. I should like to know if the activity shown on the certificate is calculated on the basis of the weight and the specific activity of the solution, or whether the activity is measured in 2 \( \pi \).

Professor Aglintsev (Translated from Russian):

In the case of first-class sample sources, their activity is measured by one of the calibration methods. Weak sources are measured on 4 \( \pi \) counters and strong sources on calorimeters. The intermediate range between those of the calorimeters and the counters can, generally speaking, be measured in ionization chambers.

In the case of control preparations, these too are measured according to the specific activity of the solutions but only for control preparations, not for sample preparations.

Dr. Grinberg (Translated from French):

Professor Aglintsev has mentioned entering on the certificate the period of the radioisotope concerned, and I should like to know if this period is the one given in the literature on the subject, i.e. an average value, one of the other values (which often differ greatly) to be found in the relevant literature, or the value determined experimentally at the Mendeleev Institute.
Other equipment

Up to now only secondary γ standardization can be performed utilizing an ion-chamber of NPL type. The laboratory is also equipped with a coincidence unit with two scintillation counters containing two $1\frac{1}{2}'' \times 1''$ NaI (Tl) crystals in connexion with EMI 6097 B photomultipliers. One of the NaI crystals can be replaced by an anthracene crystal $1\frac{1}{2}'' \times 1$ mm for β detection.

The survey given above is a summary of work performed between 1957 and 1959, which is described in full in the following reports:

2) Andersson, Ingvar Ö.: Bestämning av tritiumaktivitet i vatten. AEF-81 (1957).
   Absolutmätning av gasers aktivitet. AEF-82 (1957).
3) Martinsson, Kerstin: Absolute measurements with a $4\pi$ counter. AE-5 (1958).
   Undersökning av några faktorer som påverkar mätning i $2\pi$ räknare. RSA-21 (1959).

Only AE-5 is available for external circulation, but single copies of the other reports (written in Swedish) may be obtained from the authors.
that has been constructed. Generally speaking, I would prefer to stay within the area covered by my paper.

**Dr. Yaffe:**

I would be very glad to describe this counter to Professor Aglintsev some time, if he would like.

**Dr. Trott:**

Could Dr. Mann or Dr. Campion say what method is now preferred at the National Bureau of Standards and Atomic Energy of Canada for K-capture nuclide standardization, e.g., Fe\(^{55}\), Cr\(^{51}\)?

**Dr. Mann:**

I regret to say that we have not yet really gone into the field of standardizing electron-capture nuclides. We are thinking of trying to put Fe\(^{55}\) into an internal gas counter in the form of, I think, iron carbonyl or some such gaseous compound, which is rather corrosive. This is the approach that we are at the moment thinking of making. We have not yet tried the approach of Dr. Allen from Harwell, which has been very successful, of \(4\pi\beta\) chambers at high pressure, and I think perhaps Dr. Allen has done more in this field than anyone else at present, apart from Atomic Energy of Canada. It may be a good thing to ask Dr. Allen also to make some comments on this question.

**Dr. Allen:**

So far as I am concerned, for Fe\(^{55}\) the preferred method would be the high-pressure \(4\pi\) proportional counter, but I shall have rather more to say about that on Friday morning. For Cr\(^{51}\), I myself would prefer X-ray-\(\gamma\) coincidence counting, since it is not necessary to know the branching ratio of the \(\gamma\)-rays in order to get an absolute answer.

**Mr. Spernol (European Atomic Energy Community):**

Could Dr. Campion give more details on the colloidal silica used as a wetting agent, especially its precise name, availability, the concentration in which it is used and any significant experience acquired e.g., in connexion with measurements of S\(^{35}\), Co\(^{60}\), etc.?

**Dr. Campion:**

I have a slide here which I would like to show. The commercial supplier is DuPont in America and it is sold under the trade name of Ludox SM. According to this manufacturer's data, the silica particles are spherical, very uniform and about 60 Å in diameter. We have found that a working solution diluted 1 in \(10^4\) is a suitable solution to use. The exact amount is preferably determined by experiment and this slide will show the effect on a \(4\pi\) counting rate as a function of the amount of this diluted solution added to the aliquot. The curve shows a sharp rise to a fairly broad plateau which then gradually drops as more material is added to the source. On the left are results for S\(^{35}\) and the abscissa is in microlitres of 1 : \(10^4\) Ludox SM solution. On the right are the results for Co\(^{60}\).
mentioned, i.e., γ-ray spectroscopy, β-ray absorption and α spectroscopy — in the case of α emitters which are, of course, perhaps not very important in the radioisotopes field — radiochemical separation is quite frequently used in testing purity of radioisotope shipments. In fact, without specific separation methods for impurities, either expected or unexpected, I do not believe that we would have adequate knowledge of the purity of the isotopes being shipped. For example, the Co$^{60}$ purified preparations, as distinguished from merely irradiated cobalt, are tested for iron impurity by ether extraction — a very simple method of separation of radioiron from radiocobalt — and conversely, and much more importantly, in fact, the radioiron preparations are tested for cobalt by extracting away the iron by searching for Co$^{60}$ or Co$^{58}$ in the residue.

The Chairman:

I think we should begin discussion on purity now. I think we all know that radiochemical purity is the major possible source of errors and that we should be careful of this.

Professor Aten:

Can Dr. Mann say whether special precautions are considered necessary in the preparation of iodine samples for β counting?

Dr. Mann:

I think I might say I agree with Dr. Robinson in this case. As Dr. Robinson mentioned in his paper, we should take precautions against evaporation and so forth. In precipitating the source as a silver iodide, you have to take special precautions against evaporation.

Dr. Gross (Brazil):

In his paper Mr. Geiger described an aluminium-lined lead ionization chamber and stated that it discriminated against low-energy γ-rays and reduced the photo-electrons from the lead. Can he enlarge on this statement with regard to the complicated behaviour due to transmission of photo-electrons, the exclusion of secondary γ quanta, the percentage of transmitted photo-electrons, the Compton electron production, and the absorption rate in the walls?

What is the efficiency of the chamber? He also mentioned insets of lucite to obviate regions of reduced electronic field in the 4π counter. How does this process work?

Dr. Geiger (Canada):

It is quite correct that the behaviour is complicated, but our use of the lead-shielded aluminium chamber is based on some experiments carried out some years ago by Caster. By general experimentation on pure lead chambers, pure aluminium chambers and certain mixtures, it has been found that for radium — and I repeat this only applies to radium — the absorption corrections for the vessel containing the radium are the smallest. In brief, then, this is based on experiments and we have not really attempted to study the question theoretically.
Mr. Bučina (Czechoslovakia) (Translated from Russian):

Would Dr. Mann please indicate the size of the counter for “formamid counting”, with particular reference to the total diameter and the diameter of the liquid container and to the length and slope of the plateau of this counter.

Dr. Mann:

I think the exact dimensions of the cell were given on the slide, but I remember it was 1.1 mm deep and it is of the order of a couple of centimetres in diameter, that is the cell itself. It is cut to receive 1 ml. This is an ordinary commercial $2\pi\beta$ counting unit, the diameter of which is of the order of $2-2\frac{1}{2}$". I am just speaking from memory. The length of the plateau is something like 300 or 400 V. I do not remember the slopes, but for relative measurements it is perfectly good to, say, about 1 or $2\%$.

Dr. Pate (United States of America):

I am certainly not the only one to discover that I had missed some of the Russian publications to which Professor Aglintsev referred in his paper. Would he be so kind as to give the references concerned or to say whether reprints could be obtained through him?

Professor Aglintsev (Translated from Russian):

They have been published over the past four years in Atomnaya Energia (Atomic Energy) in 1956 and then in the Proceedings of the Institute of Metrology, issues 30/90 and 33/93, both of which appeared in 1957; and finally a symposium entitled Atomic Energy for Peaceful Purposes was published by the Power Publishing House.

If the proceedings of a conference are published, detailed references to the literature will be given in the “References” sections of the papers.

Dr. Allen (United Kingdom):

I am sorry to refer back to the question of purity again, but I would like to make a point about Cs$^{137}$. In recent years, the quantity of Cs$^{134}$ present in Cs$^{137}$ has slowly been increasing. This is presumably due to the fact that much of the old stock of fission products has now been used up and a new stock is being drawn on. I wonder if it would not be a good thing if some of the isotope production authorities produced some Cs$^{137}$ which was nearly free from Cs$^{134}$ by means of very short irradiation of uranium. We have done this on a very small scale at Harwell. I wonder whether the meeting thinks it would be a good thing to get a large quantity of this for international comparison.

Dr. Trott (United Kingdom):

I would certainly like to support that suggestion. In the Advisory Committee of the United Kingdom National Physical Laboratory, we have frequently discussed the possibility of circulating Cs$^{137}$ standards, and we have never yet done this, for the reason that Dr. Allen mentioned, that
obtained a value of 4.0 years. As you know, there was a definite cleavage between the two groups of half-lives appearing in the literature. This discrepancy was due to radiochemical instability. Tl\(^{204}\) was used in liquid solution; the solution standard was probably being absorbed to the glass, so that we were losing material and it was giving apparently a low half-life. Our approach is to check first of all on radiochemical purity by trying to verify half-lives, and then on radiochemical stability by opening our standards or using them in the conditions under which we expect the users to use them.

Dr. B. M. W. Robinson:

I think my answer to the question would be almost identical with that of Dr. Mann. We have no particular check on the radiochemistry of our solutions, which we normally receive from the Atomic Energy Research Establishment. We have had trouble in the past on occasions, when of course we have consulted sources of chemical information and sometimes started again. Thallium is a case in point which has given this kind of trouble. I think it would be more appropriate for Dr. Putman to say a little about this, as he is the origine of the standard solutions which we normally calibrate.

Dr. Putman (United Kingdom):

I would like to modify that by saying that I am near the origin of this solution, which I used to calibrate. Very careful checks of the nature described by Professor Aglintsev are done before radioactive materials are produced on a routine basis in the reactors, so that a good idea of the radiochemical impurities to be expected is obtained before the material is issued at all.

Generally, before standards are required, some experience has been gained in the production of these materials for research purposes in the reactor and so the impurities are well established. In special cases where unusual nuclides are requested for standardization, we do make special investigations which include \(\gamma\)-ray spectrometry, but not \(\beta\)-ray spectrometry.

Dr. Grinberg (Translated from French):

The reason I put my question is that we are interested in this matter and are approximately in the situation just described by Dr. Robinson. However, we are carrying on systematic \(\gamma\) spectrometry and are at present putting in two installations for checking on the basis of half-life.

Dr. Mann:

Unfortunately, Mr. Chairman, I preceded Dr. Robinson, but I too would like to say the same as he did. I notice that Dr. Reynolds, who supplies our materials from Oak Ridge, is also here now, and he might like to join Dr. Putman in answering the question.

Dr. Reynolds (United States of America):

I would like to correct the impression that we supply the materials. The materials are supplied by the Isotopes Division of Oak Ridge National Laboratory and the Atomic Energy Commission and we analyse them. The methods of analysis I can mention briefly. In addition to the methods which have been
Professor Bochkarev (Union of Soviet Socialist Republics) (Translated from Russian):

Dr. Grinberg has shown such interest in our paper by putting a number of questions that it would be simply churlish not to respond by asking him at least one question. I limit myself to one, however, as in view of the lack of time it would be inconsiderate to the other participants to put too many.

Dr. Grinberg spoke of a metal-well chamber for comparative \( \gamma \) measurements. I should be very grateful if he would explain what happens with regard to hardness in the chambers when measuring \( \gamma \) emitters of various energies.

Dr. Grinberg (Translated from French): I am afraid I do not quite understand the question.

Professor Bochkarev (Translated from Russian):

The metal-well chamber is intended for comparative measurements of \( \gamma \) sources, i.e. for measurements made by comparing various sources with a sample \( \gamma \)-ray emitter; consequently, the readings in the chamber must to some extent depend on the \( \gamma \)-ray energy. I am therefore interested to know exactly how the hardness question is dealt with, i.e. what is the dependence of the readings in the chamber on the \( \gamma \)-ray energy, and how is it taken into account.

Dr. Grinberg (Translated from French):

If you will allow me, I shall ask Mr. Engelmann, who devised this chamber, to be good enough to reply.

Dr. Engelmann (France) (Translated from French):

Actually, we have not paid very much attention to energy dependence, in view of the fact that calibrations have been made for each radioisotope. Now that we have these various calibrations we thought it would be useful to try to crosscheck them theoretically. We found that the theoretical calculations coincided fairly well with the experimental results and that the plots fell fairly well on the curve, so that it would when necessary be possible to make calibrations for nuclides whose energies are in the neighbourhood of those of other radioisotopes.

Professor Aglintsev (Translated from Russian):

May I supplement one of my replies? I should add on the question of publications that a description of electron-capture measurements was published in the journal *Experimental Apparatus and Techniques* in 1959.

Dr. Pate:

May I ask Professor Aglintsev whether, if one writes a nice letter to him in Moscow, one can obtain reprints of these publications to which he is referring?
Professor Aglintsev (Translated from Russian):

Perhaps the copies were simply distributed and one were kept, but I am trying to find out.

Dr. Yaffe:

I have a question to Mr. Braun regarding his paper. This question is regarding the wall effect which he mentioned in connexion with his gas counters. I would be interested in knowing the actual magnitude, that is as a percentage of the disintegration rate. I would have expected the wall effect to be exceedingly small, because even for an electron which originates near the wall, the back-scattering effect, combined with the gas multiplication, even though it is small near the wall, ought to be great enough to reduce this, I would have thought, to virtually insignificant proportions.

Dr. Braun (Sweden):

As a matter of fact, I have all the figures with me for the measurements on which I based the figures I gave in the paper, and if Dr. Yaffe would like to see it we can discuss it afterwards as I have the paper with me. It is quite a lengthy one.

Mr. Gasström (International Atomic Energy Agency):

I have no question, but when I heard Dr. Manov talk of time measurements, it occurred to me that what I have to say, although it is a trivial thing, might be of interest and save somebody money. We made use at the Physics Laboratory in Helsinki of the Seismographic Institute’s time piece — that is one-second sweep clock. We just attached a small mirror to it and, with the help of a lamp and a photo-cell, we were able to get second pulses which were not only very accurate to start with but also checked. This is a very cheap solution and it is quite accurate, because it is checked usually twice a day.
we have never felt that the radiochemical purity was good enough. Never-
theless at the Institute of Cancer Research, because of the practical radi-
ological interest in it, we felt compelled to have a shot at doing measurements
and estimating the Cs\textsuperscript{134} content, and this was by no means easy, even using
good multi-channel analysers and so on. In that connexion I was thinking
during the discussion that, with regard to other nuclides, it might be a very
good thing if definite special efforts were made in the case of the longer-lived
ones to obtain some stocks which were really good from the radiochemical
point of view.

**Dr. Yaffe:**

There is another problem with Cs\textsuperscript{137} and that is in the actual decay
scheme. The original 8\% has now gone down towards 3\%; I do not think this
point has ever been firmly settled and I think that some work along these
lines would be very valuable as far as Cs\textsuperscript{137} is concerned.

**Dr. Baptista (Portugal):**

I should like some information from Professor Aglintsev about a single-
wire 4 π counter. I saw a picture of it in a publication by the United States
National Academy of Sciences. The reason I ask this question is that I built
a single-wire counter and I intended to build a 4 π one, but never succeeded,
and it seems to me that was due to some polarization effects in the plastic
source-mount. I do not know if you have some information about this point
you can give me.

**Professor Aglintsev:**

What article?

**Dr. Baptista:**

The problem is that I do not know the Russian paper — only the
American reference to the Russian paper.

**Dr. Yaffe:**

I think I am responsible for this Russian reference creeping into the
American literature. At the first United Nations International Conference on
the Peaceful Uses of Atomic Energy (Geneva, 1955) there was a very lovely
counter on show at the Russian exhibit. This counter, the exhibitor told me,
was a 4 π counter which had 100 per cent efficiency. I could obtain no further
information there, and at the conference held at Easter I merely raised this
question to find out whether anyone else had seen it; then Dr. Mann, I think,
very kindly had the picture put in the proceedings, and this is how this got
into the American literature.

**Professor Aglintsev:**

I would have some difficulty in replying, as I do not exactly know
which paper is involved. It may be that you are thinking of a spark counter
II.

NEW DEVELOPMENTS OF ABSOLUTE MEASURING METHODS FOR THE STANDARDIZATION OF RADIONUCLIDES
II. 1. α-EMITTING NUCLIDES
(SESSION 3, 15 OCTOBER 1959)

II. 1.1. A NEW DESIGN FOR A HIGH PRECISION, HIGH GEOMETRY α COUNTER
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Abstract
A relatively high geometry α chamber is described which permits precision measurements on α sources deposited on a packing plate. The acceptance angle is chosen so that back-scattered particles are excluded, and this limits the geometry to not more than approximately π steradians. For the purpose of this paper, "high geometry" is defined as a useful solid angle approaching π.

A conventional counter configuration of such transmission would suffer in precision because of the large uncertainty in geometry caused by very small variations in axial position of the source. To overcome this defect a design is described in which a central axial stop is placed in such a manner that compensation is incorporated to reduce the effects on geometry of a lateral translation of the source, or use of an extended source.

Nouveau modèle de compteur α, à grande géométrie, de haute précision

Ce mémoire décrit une chambre α à géométrie relativement grande qui permet des mesures de précision sur les sources α placées sur une plaque support. L’angle d’incidence est choisi de manière à exclure les particules rétrodiffusées, ce qui limite la géométrie à environ π stéradians au maximum. Aux fins du présent mémoire, on entend par “grande géométrie” un angle solide utile voisin de π.

Si dans un compteur classique on adoptait une telle disposition géométrique, ce serait au détriment de la précision, car de très petites variations de la position axiale de la source sont la cause de graves incertitudes concernant la géométrie de la chambre. Pour pallier cet inconvénient, les auteurs proposent un modèle comportant dans l’axe central un dispositif destiné à compenser la variation axiale dans un intervalle donné. Un autre dispositif de compensation
permets de réduire les effets sur la géométrie de la chambre d'un déplacement latéral de la source ou de l'emploi d'une source non ponctuelle.

Новая конструкция высокоточного счетчика $\alpha$-частиц высокой геометрии

Дается описание счетчика $\alpha$-частиц сравнительно высокой геометрии, позволяющего проводить точные измерения источников $\alpha$-частиц, помещенных на задней пластинке. Угол приема избран таким образом, чтобы частицы отобранного рассеивания исключались, и это ограничивает примерно до $\pi$ стерадианов. В целях настоящего документа «высокая геометрия» определяется как полезный телесный угол, приближающийся к $\pi$.

В обычном счетчике конфигурации такого прохождения была бы менее точной из-за большей неясности в геометрии в результате весьма малых изменений в положении оси источника. Чтобы преодолеть этот дефект, дается описание приспособления, в котором центральный осевой зажим устанавливается таким образом, что в соответствующих пределах достигается уравнивание для изменений в положении оси. Устанавливается дополнительное уравнивание для того, чтобы уменьшить влияние на геометрию в результате поперечного перемещения источника и использования неточечного источника.

Un nuevo modelo de contador $\alpha$ de geometría alta y gran precisión

En la memoria se describe una cámara $\alpha$ de geometría relativamente alta que permite medir con precisión la actividad de fuentes $\alpha$ depositadas sobre una placa soporte. El ángulo sólido se elige de tal modo que las partículas retrodispersadas queden excluidas, lo cual limita la geometría a $\pi$ esterorradianes como máximo. Para los fines de la memoria, se entiende por "geometría alta" un ángulo sólido útil próximo a $\pi$.

La precisión de un contador corriente de las anteriores características quedaría afectada por las considerables alteraciones geométricas que originan ligerísimas variaciones en la posición axial de la fuente. El autor describe un contador que, para remediar este defecto, lleva en el eje un tope que permite compensar las variaciones axiales. El contador dispone además de un sistema de compensación para reducir los efectos de un desplazamiento lateral de la fuente sobre la geometría o para cuando se emplee una fuente no puntiforme.

The precision measurement of $\alpha$ sources is readily accomplished by means of ordinary low-geometry counters when ample activity is available. When the amount of activity is small it is necessary to use a high geometry configuration in order to obtain good statistics in a reasonable counting time. The best arrangement from the standpoint of high efficiency is a $4\,\pi$ counter in which the source is mounted on a nearly weightless film so that essentially no $\alpha$ particles are stopped before reaching the sensitive portion of the counter chamber. This method obviously cannot be used when the source is mounted on a backing plate, which is the usual case. Under these conditions it has been customary to use a $2\,\pi$ counter, and the difficulty immediately arises that a significant fraction of the $\alpha$ particles emitted into the backing plate is scattered backward and enters the counting volume. Thus a correction becomes necessary which may be from $2-4\%$, and the uncertainty of the magnitude of the correction
reduces the precision of the measurement. To avoid the need for making such a correction, the chamber geometry can be reduced to about one π steradians. The acceptance angle is now 120° and under these conditions virtually no scattered α will be detected.

For the purpose of this paper, a high geometry counter will then be defined as one approaching π steradians in solid angle.

When a 1 π counter of convenient physical dimensions is examined, another difficulty appears, namely, that the geometry factor is very sensitive to the positioning of the source. For example, in a 1 π counter with a working distance from source to collimator of 44 mm and a collimator diameter of 154 mm, a vertical displacement of the source of only 0.1 mm will alter the geometry by 0.2%/α, and to reduce the uncertainty to 0.05%/α requires that the sample position be known to 0.025 mm, a nearly impossible requirement. Thus, some type of compensating device is required to make the geometry less sensitive to source position. One such possibility is shown in figure 1.

A circular stop is placed above the source. The geometry is reduced to $G_c - G_b$, where $G_c =$ solid angle of collimator of radius $c$, and $G_b =$ solid angle subtended by the stop of radius $b$. Since the stop is closer to the source than the collimator, $G_b$ will be relatively more sensitive to source positions than $G_c$. By suitable choice of $a$ for a given stop radius $b$, the change of geometry with source position can be made zero. By simple calculation it is found that the following relationship between $a$ and $b$ must hold for perfect compensation:

$$a = \frac{b^{2/3}}{c^{2/3}} \sqrt{c^2 + h^2 - C^{2/3} b^{2/3}}$$

An example will illustrate the compensation obtainable with such a stop. Let $h = 44$ mm, $C = 77$ mm, and $b = 11$ mm. Then $a = 21.5952$ mm, and $G = 0.19746$. Curve A in figure 2 shows how the geometry factor varies with position of source. Curve B shows the variation which would be obtained with a chamber having no stop but with $h = 58.52$ and $C = 77$ mm as before.
And now a further complication arises. The previous curves have been drawn for point sources, but the usual source is from one millimeter to several centimeters in diameter, so the effective geometry of a point off axis becomes of importance. Figure 3 plots the variation of geometry factor versus displacement of a point source of a chamber having the same dimensions as previously given. It is evident that non-uniformity is extremely serious from the standpoint of a precision counter. An obvious remedy is to use one or more additional stops above the first one and of such size as not to change the geometry of a point on the axis but to cast an additional shadow on the detector for an off-axis source. Figure 3 indicates that a correction resembling more or less a quadratic function will be required, whereas a single additional stop adds a nearly linear correction, hence it is concluded that an infinite number of such stops will be required, i.e., a figure of revolution continuous from the initial position to the plane of the collimator.
The general mathematical treatment of the problem is extremely difficult, but fortunately for practical purposes it can be simplified and solved by direct but tedious numerical methods.

It is first necessary to determine the shape of the shadow in the plane of the collimator in terms of an arbitrary shape of stop. The only figure which could be handled analytically was a conic section whose equation can be written

\[ \frac{(u-k)^2}{a^2} + \frac{v^2}{m^2} = 1 \]

Where \( v \) = radius of the stop measured a distance \( u \) from the bottom end, and \( k, a^2, \) and \( m^2 \) are arbitrary constants (see figure 4). If \( m^2 \) is positive the figure is an ellipse, if negative it is a hyperbola. Introducing two required conditions reduces the equation to

\[ \frac{v^2}{b^2} = \frac{b^2}{m^2-b^2} \cdot \frac{u^2}{a^2} + 2 \frac{u}{a} + 1 \]

As before, \( a \) = optimum distance from stop to source, \( b \) = radius of bottom end of stop, and \( m^2 \) is the only parameter remaining to adjust for best performance. The shadow cast by this stop from an off-axis point is a semicircle of radius \( \frac{bh}{a} \) joined by half an ellipse, with semi diameters of \( \frac{bh}{a} \) and

\[ \frac{bh}{a} \sqrt{1 + r^2 \left( \frac{2}{b^2} - \frac{1}{m^2} \right)} \]

where \( r \) = distance of source from the axis.

On the basis of numerical integrations carried out on the shadow a stop was made with \( m^2 = 2 a^2 \). To avoid scattering off the surface and to simplify the fabrication it is made with a number of steps, as shown in figure 5. These steps introduce a slight error, which varies approximately as the cube of the horizontal displacement of the source, and inversely as the square of the number of steps. With a source 20 mm off axis the error is less than 0.02% if the steps are about 2.5 mm as in the present case.
Figures 6 and 7 show a cross-section drawing and photograph of the counter as finally constructed. The collimator diameter is 152.405 mm, stop diameter 21.77 mm at bottom end, the distance from source to stop, $\alpha = 21.37$ mm, and from collimator to bottom of stop 22.17 mm. The calculated geometry factor is 0.19748. A zinc sulfide-Ag screen is deposited...
on the glass plate and scintillations from the alpha particles are detected by a photomultiplier tube above the glass plate (2).

Figure 7
Photograph of counter.

The glass plate is 12.6 mm thick and under vacuum deflects 0.012 mm. This was allowed for in making the stop.

The performance of the counter was tested using an Am^{241} source about 2 mm diameter on platinum, and the count rate plotted as a function of source displacement both vertically and horizontally.

In figure 8 the straight line is the theoretical variation for an uncompensated chamber having the same geometry factor and collimator diameter. The curve is the theoretical performance of a chamber having a single plane circular stop. A solid stop will cause the curve to fall off more rapidly on the "down" side, but will not alter the "up" half.

Figure 8
Observed counting-rate versus vertical displacement.
Figure 9 presents the preliminary data taken with this counter showing performance as a function of horizontal displacement of source at several values of \( h \), indicated by values of \( \Delta h \) above or below the design value. Also shown are calculated values for an uncompensated 1\( \pi \) chamber and a chamber having the same geometry as at present but with only a single plane stop compensator.

Each point in both figures represents from two to ten million counts (2 to \( 10 \times 10^6 \)), so that the standard deviation of each point is about 0.05% on the average.

On the basis of the results obtained so far it is concluded that the design is sound and the counter is successfully operating. It may be possible to remove the dip in the curves of figure 9 by slightly altering the shape of the elliptical stop, probably in the upper half. It should be noted that ordinarily the error in locating the vertical position of the source will not be more than 0.1 mm, so that there need be no concern about the divergence of the curves plotted in figure 9. The two closer curves represent a relative displacement of 0.33 mm.

A single cross-check of the geometry of this chamber with a precision medium-geometry counter gave agreement within 0.03%.

In the future the effect of making \( \alpha \) smaller will be investigated. It may be possible by this means to extend the flat portion of the curves shown in figure 9, although for present purposes the performance is entirely satisfactory.

This paper describes one means of compensating a counter to make its effective solid angle independent of source position, within limits.

A second method could utilize a series of collimators above or below the primary one, and a combination of both methods could probably be used to obtain horizontal compensation to appreciably greater distances than at present.

REFERENCES

(1) JAFFEY, A. H., Private communication.

II. 1.2. THE DETERMINATION OF THE HALF-LIFE OF U\textsuperscript{238} BY ABSOLUTE COUNTING OF \( \alpha \) PARTICLES IN A 4 \( \pi \)-LIQUID SCINTILLATION COUNTER

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Abstract

The specific activity of natural uranium was determined by liquid scintillation \( \alpha \)-counting. Uranium was extracted from its decay products by methyl isobutyl ketone extraction and samples of this solution were added directly to the liquid scintillator. A quantitative investigation was made of the separation of uranium from thorium by the extraction method employed. Assuming that U\textsuperscript{238} and U\textsuperscript{234} were in equilibrium, and correcting for the presence of U\textsuperscript{235}, the specific activity and the half-life of the isotope U\textsuperscript{238} were calculated.

Détermination de la période de l'U\textsuperscript{238} au moyen du comptage absolu de particules \( \alpha \) dans un compteur 4 \( \pi \) à scintillateur liquide

L'activité spécifique de l'uranium naturel est déterminée au moyen d'un comptage \( \alpha \) par scintillateur liquide. L'uranium est séparé de ses produits de désintégration par une extraction à la méthylisobutylcétoné et des échantillons de cette solution sont ajoutés directement au scintillateur liquide. On fait une étude quantitative de la séparation de l'uranium et du thorium par le procédé d'extraction utilisé. En admettant que U\textsuperscript{238} et U\textsuperscript{234} sont en équilibre et en faisant la correction voulue pour tenir compte de la présence de U\textsuperscript{235}, on calcule l'activité spécifique et la période de U\textsuperscript{238}.

Определение периода полураспада U\textsuperscript{238} посредством абсолютного счета \( \alpha \)-частиц в жидкостном сцинтилляционном счетчике 4 \( \pi \)

Специфическая активность естественного урана была определена с помощью жидкостного сцинтилляционного счетчика \( \alpha \)-частиц. Уран был получен из продуктов его распада экстракцией метил-изобутилкетона, и образцы этого раствора были добавлены непосредственно в жидкый сцинтиллятор. Было предпринято количественное исследование отделения урана из тория с помощью метода экстракции. Предположив, что U\textsuperscript{238} и U\textsuperscript{234} находятся в равновесии, и сделав поправку на присутствие U\textsuperscript{235}, были подсчитаны удельная активность изотопа U\textsuperscript{238} и период его полураспада.

Determinación del período del U\textsuperscript{238} por recuento absoluto de las partículas \( \alpha \) con un contador 4 \( \pi \) de centelleador líquido

La actividad específica del uranio natural se determinó con un contador \( \alpha \) de centelleador líquido. El uranio se separó de productos de desintegración por extracción con metilisobutilcetona, y muestras de esta solución se añadieron directamente al centelleador líquido. Se estudió cuantitativamente el grado de separación uranio/torio alcanzado con el método de extracción empleado. Introduciendo correcciones para tener en cuenta la presencia de U\textsuperscript{235}, se calculó la actividad específica y el período de semidesintegración del U\textsuperscript{238} suponiendo que este isótopo se encontraba en equilibrio con el U\textsuperscript{234}.
Introduction

Three measurements of the half-life of $^{238}$U were listed in the Table of Isotopes compiled by Strominger et al. (4). In these determinations the specific activity of $^{238}$U was found by $\alpha$ counting of thin, weighed uranium deposits in ionisation chambers having a known geometry. The method required correction of the results for back-scatter and self-absorption.

It has been shown that the $4\pi$ liquid scintillation counting of $\alpha$ particles yielded absolute disintegration rates (5) (6) (7) and this method was employed in the measurement of the specific activity of natural uranium reported here. Owing to the mono-energetic nature of the $\alpha$ particles, a well-defined peak was obtained in the corresponding impulse distribution when a sample of uranium was dissolved in the liquid scintillator. Since conditions could be attained such that this peak stood well above photomultiplier noise, the $\alpha$ particles could be counted with practically a 100% efficiency, there being hardly any geometry, and no absorption or scattering effects to be taken into account.

The extraction of uranium from its decay products

Since it was not possible to discriminate between impulses caused by $\alpha$ and $\beta$ particles in the liquid scintillation counter, the success of the counting method depended on the complete separation of uranium from its $\beta$ active daughter, $^{234}$Th. The separation method used, was the extraction of uranium with methyl isobutyl ketone ("hexone") from an aqueous solution. Since it was known that under certain conditions thorium could also be extracted by this solvent (8) (9), it was necessary to investigate the extraction of this element. Free nitric acid, and salting-out agents such as ferric and ammonium ions were found to enhance the solubility of thorium in hexone, whereas the presence of uranium had the opposite effect.

Experimental

The preparation of pure-uranium solutions was as follows:

About 10 to 20 g of analytical reagent-grade uranyl nitrate, or alternatively, a product prepared in our own laboratory from 90% uranium oxide ore, were dissolved in 10 to 20 ml of water, two drops concentrated nitric acid were added, and uranium was extracted with 50 ml hexone. The hexone phase, containing about 35% of the uranyl nitrate, was separated and divided into two parts. One part was used immediately for counting, the other for analytical determination of the uranium concentration.

The following method of chemical analyses was used:

About 10 g of chemical were weighed out accurately, the organic material removed and uranium precipitated by carbon dioxide — free ammonia. The precipitate was separated by filtration, ignited at about 900°C and weighed as $\text{U}_3\text{O}_8$. All analyses were done in duplicate and deviations from the mean were always less than 0.06%.

Completeness of precipitation was checked by a test on the filtrate, having a sensitivity conservatively estimated as better than 0.05 mg $\text{U}_3\text{O}_8$. The pre-
cipitate was afterwards redissolved and tested for impurities, of which none could be found.

In order to determine how much thorium was extracted by this procedure, some test extractions were carried out as described above on uranium having known amounts of pure thorium nitrate added to it. No thorium was found in these extractions. The sensitivity of the analytical method employed here was conservatively estimated to be better than 0.03% for that extraction where the aqueous phase contained the largest amount of thorium, namely 0.81 g of the element. From this it was deduced that a hexone extraction of uranyl nitrate slightly acidified with nitric acid would yield a uranyl nitrate solution without any observable Th$^{234}$.

For determining the specific activity two different but similar counting arrangements were used, details of which were described elsewhere (10).

The liquid scintillator consisted of a mixture of either toluene and ethyl alcohol, or toluene and π dioxane as solvent with P. P. O. * and P. O. P. O. P. ** as primary and secondary solutes, respectively. Concentrations of P. P. O. and P. O. P. O. P. were 14 grams per litre and 0.5 grams per litre respectively. Alcohol or π dioxane was necessary to ensure that the uranium stayed in solution in the scintillator, a most important requirement. Ten per cent of alcohol or 50% of π dioxane was used to achieve this.

Samples of the hexone solutions were weighed and transferred to the sample cells by means of weighing pipettes, mixed with 12 ml of liquid scintillator and counted. Counting rates varied with uranium concentration and sample size between about 4000 and 10000 counts per minute at which rates losses due to paralysis in the counting channels were insignificant.

Figure 1(a) is an oscillograph of a typical impulse distribution. The α peak had a half-width of about 40% as measured with a differential discriminator, the rays of U$^{238}$ and U$^{234}$ being unresolved. The counting rates were determined with increased amplification, so that the impulses just saturated the non-overloading amplifier, as shown in figure 1(b). The counting was then done at a number of discrimination levels below the maximum output of the

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*) ***) Supplied by Nuclear Enterprises Ltd.

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Figure 1 (a)

Impulse distribution.

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9) 99) Supplied by Nuclear Enterprises Ltd.
amplifier, but above the phototube noise. In this way a discrimination curve was obtained. Under ideal conditions the slope of such a discrimination curve should be zero. Imperfect counting conditions, and the presence of uranium daughter activities would cause the discrimination curve to have a negative slope. The occurrence of such a slope would therefore be an indication of the significance of perturbing effects and thus a measure of the reliability of the α counting.

All samples were counted within seven hours of the extraction in order to keep the contribution due to the growth of daughter activities as low as possible. Counting time was about an hour per source and eight sources were counted per extraction, using both counting arrangements simultaneously. Counting rates were corrected for background which varied between 1/2% and 3% of the true counting rates. Since the counting was done well above phototube noise the background was stable and reproducible.

Due to the growth of daughter activities an increase in counting rate occurred which was just perceptible after seven hours. In order to determine a correction for this increase, sources were recounted on the day following the measurements, when a significant increase in slope was usually also found.

Results and Discussion

Examples of the discrimination curves are shown in figure 2. Least squares analysis showed that only eight of the forty distributions obtained had slopes significantly different from zero. Furthermore in ten cases the slopes were positive. It was therefore clear that the true slopes of the curves (expected to be negative) were so small that in most cases they were completely masked by statistical variations.

The mean of the calculated slopes was found to be negative and equal to 0.46% over the range of discrimination levels where the measurements were made.
From considerations of \( \alpha \) particle range and the dimensions of the scintillating medium it was calculated that about 0.32\% of the \( \alpha \) particles penetrated beyond the surfaces of the solution and did not lose all their energy in the scintillator. This effect would cause an increase in counting rate of 0.16\% over the range of discrimination levels under consideration. The increase over the same range due to the growth of daughter activities was determined as 0.29\% for the average interval (4.1 hours) between extraction and counting. Thus, by these considerations, the value of the mean slope given above was fully explained.

![Discrimination curves](image)

The results were analysed in two ways:

1. Assuming that extrapolation to zero impulse-height was unnecessary, the counting rates obtained at different discrimination levels were regarded as repetitions and their mean taken as the counting result for each source;

2. Alternatively, the calculated intercepts on the zero impulse-height axis were taken to represent the true source activities.

Two sets of results were thus obtained and are referred to in what follows as the "means" and the "intercepts".

The average increase in disintegration rate due to the daughter activities was about 1.7\% and 3.1\% per 24 hours for the "means" and the "intercepts" respectively. Accordingly, an average subtractive correction of 0.071\% for every hour elapsed since extraction, was applied to all the "means", and a similar correction of 0.13\% to all the "intercepts".
The α activities of five hexone-uranium solutions are given in Table I.

### Table I

<table>
<thead>
<tr>
<th>Solution No.</th>
<th>Uranium Concentration, weight per cent</th>
<th>α activity of solution, disintegrations per minute per milligram</th>
<th>Specific activity of natural uranium, disintegrations per minute per milligram</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>1 &quot;means&quot;</td>
<td>“intercepts”</td>
</tr>
<tr>
<td>1</td>
<td>7.101</td>
<td>107.6640</td>
<td>107.7791</td>
</tr>
<tr>
<td>2</td>
<td>6.650</td>
<td>100.8370</td>
<td>100.9910</td>
</tr>
<tr>
<td>3</td>
<td>5.670</td>
<td>85.9268</td>
<td>86.0007</td>
</tr>
<tr>
<td>4</td>
<td>3.954</td>
<td>60.1079</td>
<td>60.1484</td>
</tr>
<tr>
<td>5</td>
<td>2.645</td>
<td>40.1729</td>
<td>40.2932</td>
</tr>
</tbody>
</table>

Specific activity of natural uranium, disintegrations per minute per milligram  
1520 ± 2

Specific activity of U²³⁸, disintegrations per minute per milligram  
747 ± 1

Half-life of U²³⁸, years  
(4.46 ± 0.01) × 10⁹

These values combined with the corresponding analytical determinations of the uranium concentrations, yielded the specific activity of natural uranium. Table I shows that the two averages obtained by "means" and "intercepts" respectively did not differ significantly. Consequently the value of 1520 ± 2 disintegrations per minute per milligram was finally taken for the specific activity of natural uranium. From this the specific activity of the isotope U²³⁸ was calculated on the assumption that U²³⁸ and U²³⁴ were in equilibrium and correcting for the presence of U²³⁵. Uncertainties in the U²³⁵ half-life used affect the final result through this correction, but not seriously, since the calculated contribution of U²³⁵ to the specific activity of normal uranium was only 2.26%. Finally the half-life of U²³⁸ was calculated from its specific activity as 
(4.46 ± 0.01) × 10⁹ years. Errors quoted are statistical only, expressed on the basis of the 95% probability level. Data used in the calculations are listed in Table II.

### Table II

<table>
<thead>
<tr>
<th>Abundance:</th>
<th>U²³⁵</th>
<th>0.72%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Half-Life:</td>
<td>U²³⁵</td>
<td>7.1 × 10⁸ years</td>
</tr>
<tr>
<td>Atomic Weight:</td>
<td>U²³⁵</td>
<td>235.117</td>
</tr>
<tr>
<td>Atomic Weight:</td>
<td>U²³⁸</td>
<td>238.125</td>
</tr>
<tr>
<td>Abundance:</td>
<td>U²³⁸</td>
<td>99.28%</td>
</tr>
<tr>
<td>Avogadro’s Number</td>
<td></td>
<td>6.02501 × 10²³</td>
</tr>
<tr>
<td>Number of minutes per year</td>
<td></td>
<td>5.2596 × 10⁵</td>
</tr>
</tbody>
</table>
$^{238}\text{U}$ half-life values listed in the Table of Isotopes are given in Table III.

<table>
<thead>
<tr>
<th>Half-life, years</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>$(4.56 \pm 0.03) \times 10^9$</td>
<td>Leachmann and Schmitt, 1956</td>
</tr>
<tr>
<td>$4.507 \times 10^9 \pm 0.2%$</td>
<td>Kovarik and Adams, 1955</td>
</tr>
<tr>
<td>$(4.49 \pm 0.01) \times 10^9$</td>
<td>Kienberger, 1949</td>
</tr>
<tr>
<td>$(4.46 \pm 0.01) \times 10^9$</td>
<td>This work</td>
</tr>
</tbody>
</table>

It is seen that the half-life determined in this work is somewhat smaller than these values. Closest agreement is with Kienberger's value where the discrepancy is 0.67%. 

**REFERENCES**


(10) STEYN, J., "The Comparative Accuracy of the 4 π-Liquid Scintillation Counting Method of Radioisotope Standardization", (see paper II. 2. 8. page 278).
II. 2. β-EMITTING NUCLIDES
(SESSIONS 3 AND 4, 15 OCTOBER 1959)

II. 2.1. DISINTEGRATION-RATE DETERMINATION
BY 4π COUNTING

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Abstract

The history of the development of the 4π counting method is reviewed. The unique properties of the 4π counter system are described and the criteria are specified which place disintegration-rate measurements, made with the system, on an absolute basis. The types of radioactive decay process to which the method is applicable are outlined, together with the various errors to which a given measurement is liable. Departure of response probability from unity, absorption of radiation in the source-mounting system, and source self-absorption are discussed in detail.

Détermination du taux de désintégrations par comptage 4π

L'auteur fait l'historique des progrès de la méthode de comptage 4π. Il décrit les propriétés remarquables du dispositif de comptage 4π et précise les conditions dans lesquelles les mesures du taux de désintégration se font avec ce dispositif sur une base absolue. Il donne un aperçu des types de processus de désintégration radioactive, ainsi que de diverses erreurs auxquelles est sujette une mesure donnée. Il examine en détail le cas où la probabilité de réponse s'écarte de l'unité, l'absorption des rayonnements dans le support de la source et l'autoabsorption de la source.

Определение скорости распада при помощи 4π счетчиков

Дается обзор истории развития метода счета с помощью 4π счетчиков. Приводится описание уникальных свойств системы 4π счетчиков, уста-
Determinación de la velocidad de desintegración por recuento $4\pi$

En la memoria el autor expone el desarrollo del método de recuento $4\pi$. Describe las características particulares de este método y explica las razones por las que las mediciones de la velocidad de desintegración efectuadas según ese procedimiento tienen un carácter absoluto. Se indican, asimismo, los tipos de procesos de desintegración radiactiva a los que se puede aplicar el método, así como los diferentes defectos de que puede adolecer una medición determinada. También se estudia detenidamente el caso en que la probabilidad de respuesta no es igual a la unidad, la absorción de radiaciones en el soporte de la fuente y la autoabsorción de la misma.

This is intended to be a review paper covering the work on the $4\pi$ counting method published up to the present time. It must, therefore, necessarily rely on data which have been presented before; it is to be hoped, however, that some of them will be new to some of the audience, and that repetitious detail can be avoided.

$4\pi\beta$ counting is the most widely used nuclide-standardization method. And yet we have already seen at this conference that, despite all the advances in technique that have been made, all is not well. Disagreement among disintegration-rate values from various laboratories may be of several per cent, even with nuclides that should be favorable ones to measure. It seems timely then to review the method. Let us observe which points we agree upon, or think we understand, and those upon which uncertainty still exists. This will clear the way for later papers in this session which will deal with these latter aspects.

First, however, one possibly should comment on the light shed upon the precision and accuracy of particular standardization methods by inter-laboratory comparison of solution standards. Let us neglect effects resulting from the manner in which samples are circulated, which may possibly be responsible for the disagreements in some cases. Then the fact that there is still a scatter in the results reported by different workers for a given standard cannot be taken as necessarily reflecting on the capabilities of the method used. $4\pi$ counting, for example, is very much what you make it. Many of the techniques to be used are certainly difficult ones, and failures to duplicate some observations or performance can, undoubtedly, be attributed to this. In addition, it is difficult to draw conclusions from intercomparison results unless copious details are simultaneously available as to the exact techniques used. It is to be hoped that such could be arranged in future intercomparisons.
A brief history of $4\pi$ counting is contained in figure 1 in which a selection of the contributors who have published experimental work on this subject are listed with the date of their first major publication. The bibliography at the end of this article is substantially complete with respect to published work on the $4\pi$ counting method. It is interesting to observe that a continuous flow of contributions has appeared in the last decade, and has originated from many widely separated laboratories in many countries.

J. A. Simpson 1944
L. Meyer-Schützmeister
(O. Haxel and F. G. Houtermans) 1948
R. Cohen 1949
J. R. Beyster and M. L. Wiedenbeck 1950
G. A. Sawyer and M. L. Wiedenbeck 1950
C. J. Borkowski 1950
G. Charpak and F. Suzor 1950
R. C. Hawkings, W. F. Merritt and J. H. Craven 1951
H. H. Seliger and L. Cavallo 1951
F. G. Houtermans, L. Meyer-Schützmeister and D. H. Vincent 1952
W. B. Mann and H. H. Seliger 1953
D. B. Smith 1953
B. D. Pate and L. Yaffe 1955
P. J. Campion 1957
Y. Le Gallic and M. Thenard 1957
K. Martinsson 1958
R. Gunnink, L. J. Colby Jr. and J. W. Cobble 1959
G. Bertolini and O. Terracini 1959

Figure 1
Experimental contributors to the field of $4\pi$-counting.

A review paper should reiterate the well-known advantage of a counting system which subtends a solid angle of $4\pi$ steradians at a sample of radioactive material. If the counter system can be made to respond to every charged particle leaving the source, then one count will be registered per disintegration, provided the source emits with unit probability one charged particle per disintegration.

This simple result is undisturbed by emission of electromagnetic radiation, auger or conversion electrons or by scattering of radiation within the counter system, providing these events occur within the resolution time of the system, which can be provided for experimentally. The contrast with lower geometry counting systems, in which these secondary effects are major sources of error, is obvious.

It is the elegance of the counting-rate: disintegration-rate relation and the simplicity of the apparatus used that make the $4\pi$ counting method, at least in ideality, a recommended standardization method (35) for negative $\beta$-particle emitters and $\alpha$ emitters. It is indeed one of the few absolute methods available for materials which emit only $\beta$ radiation. In the case of nuclides decaying partly or wholly by electron capture, however, it is not an absolute method in the same sense, since decay scheme data are required for interpretation of results.
Experimentally, the accuracy with which disintegration-rate measurements can be made depends on two factors:

1) how nearly one can bring the counter to responding to every charged particle emerging from the source
2) how successful one can be in preparing the radioactive sample so that every charged particle emitted emerges into the counter system. Let us discuss each briefly:

1) A counter system subtending $4\pi$ steradians at the source can be made in several ways; most work has been done with GM or gas-filled proportional counters. The design of chamber is nearly always two identical $2\pi$ counters facing each other with the source between them on a grounded mounting foil. Figure 2 is a diagram of a chamber published by Hawkins et al. (16). A variety of spherical and cylindrical configurations have been successfully employed; an important feature is apparently a region of high field-strength in the counter of the chamber in the vicinity of the source.

The two halves of the counter are operated with their anodes connected in parallel. Operation with the two half-counters disconnected leads to the observation of several strange phenomena, including half-counter counting-rates greater than half the connected $4\pi$ counter rate, and signals of positive polarity in addition to the usual negative signals from electron collection. There is even some international disagreement as to the interpretation of some of these effects. However, this is all irrelevant, in the context of disintegration-
rate determination, for which (one repeats) the two anodes are connected in parallel.

There is a certain philosophical question that now arises. Even if one is employing a counter geometry which subtends essentially 4π steradians at the source, how is one to establish that every charged particle entering the counter gas is registered? A recent publication (36) states:

"In the last analysis, the accuracy of the 4π counter depends largely upon

![Figure 3](image.png)

Counting-rate vs. high-voltage curves for beta minus emitters with a range of end-point energies.

using as the assay the highest reproducible possible counting-rate that has been obtained after many trials. This is a highly unsatisfactory basis of standardization."

Fortunately the situation can be somewhat better defined. Since the sensitive volume of a 4π counter surrounds the source, a properly designed proportional counter system, in which the maximum field strength in the counter is at the source-position, yields very long flat counting-rate/high-voltage plateaus. Published data (27) (38) have shown counting-rates of α- and β-emitting sources to remain constant to ~ 0.1% when the high voltage was raised as much as 1000 volts from a starting voltage of 2000 or 3000 volts, corresponding to an increase of gas-gain of two orders of magnitude. Figure 3 shows typical plateaus with β emitters of increasing end-point energy. We illustrate with our own data (27), but similar data have been published several times. Cautious workers also investigate the effect of variation of other system parameters, e. g. bias voltages,
Absorption of beta radiation in the source mount.

$NT$ — the ratio of counting-rate to disintegration-rate with the source mounted on films of increasing thickness.

$Ns$ — the ratio when the source is sandwiched between two films of this thickness.

$NA, N^{A}$ — apparent disintegration-rate calculated from the sandwich data by two published procedures (16, 12 respectively).

diameter of aperture over which the film is mounted, source diameter, etc., and in favorable cases show them to have similarly small effect on the counting-rate. Also counter-system dead-time losses are preferably determined empirically by the multiple-source method or some similar technique, rather than calculated from the usual (and often unreliable) relationships.
In addition, a direct measurement of system parameters, such as gas multiplication and amplifier gains, collector system capacitances and bias voltages, allows one to calculate, at some voltage within the operating range, the minimum amount of ionization in the counter gas that will cause an event to be registered. Values of 1 to 8 ion-pairs have been reported \(^{(27)}\) \(^{(38)}\) which by statistical calculation can be shown to correspond to registration of better than 99.9\% of the \(\beta\) particles from \(^{36}\)S for example. Thus the response of a properly designed proportional counter to \(\beta\) particles can be shown in a substantial way to be satisfactory, and to \(\alpha\) particles it is equally good. Likewise, GM counters, sensitive to one ion-pair and free from spurious-pulse effects, may be shown to have a similar satisfactory response by similar arguments.

2) The other major problem is to ensure that as great a fraction as possible of the radiation to be measured emerge into the counter gas, and that the fraction that does not emerge be determined.

The problem of fabrication of films strong enough to act as satisfactory mounting media for the source in a 4π counter, and determination of their transmission to various radiations has effectively been solved. Films of synthetic resins can be made by published methods \(^{(7)}\) \(^{(16)}\) \(^{(17)}\) \(^{(21)}\) \(^{(22)}\) \(^{(26)}\) thin enough so that their absorption of \(\alpha\) radiation is quite negligible, and of \(\beta\) radiation is either negligible or, in the case of the weakest energy radiation, can be corrected for. Figure 4 is typical of published data \(^{(17)}\) \(^{(19)}\) \(^{(22)}\) \(^{(28)}\) allowing correction for absorption of radiation in the source mount. Some disagreement still exists on matters of detail, such as the thickness of metal that must be vaporized onto the films to ground the sources, figures of between 2 and 30 \(\mu\)g/cm\(^2\) of Au for example have been found \(^{(16)}\) \(^{(27)}\) \(^{(34)}\) \(^{(38)}\), but the major problems are now solved, as shown by the agreement among the independent data in figure 5, due to MARTINSSON \(^{(37)}\) and added to by us. It should be noted that the data in column headed NPL, were obtained by the “sandwich” method \(^{(16)}\) the defects of which have been discussed \(^{(28)}\) \(^{(34)}\).

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>20 (\mu)g/cm(^2) Plastic Film</th>
<th>200—220 (\mu)g/cm(^2) Al or Plastic</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Martinsson Pate &amp; Yaffe</td>
<td>Martinsson Pate &amp; Yaffe</td>
</tr>
<tr>
<td>(^{32})P</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>(^{124})Na</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>(^{198})Au</td>
<td>0.2</td>
<td>&lt;0.1</td>
</tr>
<tr>
<td>(^{204})Tl</td>
<td>0.3</td>
<td>0.2</td>
</tr>
<tr>
<td>(^{35})Se—(^{90})Y</td>
<td>0.2</td>
<td>0.1</td>
</tr>
<tr>
<td>(^{31})I</td>
<td>0.2</td>
<td>0.4</td>
</tr>
<tr>
<td>(^{60})Co</td>
<td>0.5</td>
<td>0.6</td>
</tr>
<tr>
<td>(^{45})Ca</td>
<td>1.0</td>
<td>1.0</td>
</tr>
</tbody>
</table>

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</tr>
<tr>
<td>(^{45})Ca</td>
<td>1.0</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Figure 5

Correction for 4π counter source-mount absorption for various \(\beta\) emitters.
The same happy situation does not exist in the problem of self-absorption. The status of $4\pi$ counting as a method of standardization is at present decided on the self-absorption question. The other widely used method of $\beta$-emitter standardization, the coincidence method (which includes $4\pi$ $\beta$-$\gamma$ coincidence counting), is much less troubled by self-absorption of $\beta$ radiation. This method does, however, have problems of its own, and indeed is inapplicable to a number of nuclides of interest such as those that emit only $\beta$ radiation.

Self-absorption effects in $4\pi$ counters with $\alpha$ emitters and with $\beta$ emitters of end-point energy greater than 1 MeV should be small enough to be neglected with the source weights usually encountered in standardization work. They become quite troublesome when the energy is less than 0.5 MeV, and have received most attention in connection with standardization of $S^{35}$ and Co$^{60}$. Progress towards solution of the problem has, however, been made.

Figure 6
Phase-contrast photomicrograph ($\times$ 20) of a deposit from the evaporation of Na$_2$SO$_4$ solution under infra-red irradiation on 5 $\mu$g/cm$^2$ VYNS film without insulin.
Investigation of methods of mounting source materials in a fashion which would render self-absorption effects minimal and reproducible has pursued two main lines. In the first, deposition of material from solution has been studied, since solution aliquots are a convenient means of sampling radionuclides, usually supplied in solution form. Deposition has been accom-

![Figure 7](image)

**Figure 7**
Phase-contrast photomicrograph (×20) of a deposit from the evaporation of Na$_2$SO$_4$ solution under infrared irradiation on 5 µg/cm$^2$ VYNS film after pretreatment of the film with 5% insulin solution.

plished by evaporation of the solution droplet on the source mounting film, although this may lead to local aggregation of material on dust particles or at the periphery of the area originally covered by the droplet, as in figure 6. Some workers prefer aluminium foil source-mounts, despite their necessarily greater thickness than plastic films and inferior chemical properties, because of the superior evenness of evaporation deposits obtained. Others have used plastic films, pretreated with various wetting agents, of which insulin in various forms seems the most successful. The type of deposit so obtained is shown in figure 7. The successful preparation of $^{35}$S sources by this means has been reported, and also sources of Ni$^{63}$ ($\beta$ end-point energy 70 keV). Alter-
natively, as often used for I$^{131}$ or Co$^{60}$, the sample may first be precipitated from solution. Reports indicate, however, that uneven deposits are often obtained by this method (25). We also heard yesterday of the use of colloidal silica to improve the evenness of deposits.

As an example of a different approach, preparation of sources by vaporization of a suitable chemical compound on to the source mount in vacuo has been reported (10) (30) and an electron micrograph of a deposit so obtained is shown in figure 8. These crystals are of 1 micron diameter. This method was first applied to alkali halides, and its use for a number of nuclides will be described in another paper at this meeting.*

Figure 8
Electron micrograph (X 10000) of a deposit of cobalt 1-nitroso-2-naphthol on VYNS film. Deposit, produced by vacuum distillation, was of 1.2 µg/cm² superficial density.

The determination of the extent of self-absorption in a given sample has been effected in two ways. Firstly, if a series of sources of the same nuclide can be produced in a reproducible fashion from solutions of increasing dilution (16), then it may be possible say to extrapolate to infinite dilution and thus to assess the effects of self-absorption. However, reduction of the weight of

* Paper II. 2. 2.
material in a source does not necessarily reduce its average superficial density. Evaporation of dilute solutions can lead to local but invisible aggregates formed, for example, around dust particles, and further dilution of the initial solution will increase the spacing between such aggregates without necessarily affecting their size. So self-absorption effects may still be important and unsuspected.

On the other hand, one can increase both the weight of the source and the self-absorption (30) and determine the latter from an extrapolation of a plot of specific activity as source thickness in the conventional manner, as will be discussed in another paper.

An alternative approach is to incorporate into a source of a radionuclide, prepared for \( {\mathcal{4}}\pi \) counting, a quantity of a second nuclide with similar \( \beta \) energy but one which is assayed by an alternative means, such as the \( {\mathcal{4}}\pi \beta-\gamma \) coincidence method (34). One then uses the self-absorption data for the additive applied to the nuclide to be assayed in a manner to be discussed in a later paper. Alternatively, one can determine self-absorption coefficients for a variety of materials and \( \beta \) energies and apply these to other sources prepared later (36). This relies heavily on the reproducibility of one's technique for source fabrication.

Thus it can be seen that the application of \( {\mathcal{4}}\pi \) counting with accuracy to the assay of low-energy \( \beta \) emitters is a difficult problem in analytical chemistry. Each assay is really a research problem in itself. This is a price that must be paid for the simplicity and elegance of the method, and its relative freedom from decay scheme assumptions and other corrections.

For the assay of some nuclides one really has little choice but to pay this price, for others the price is low enough. For still others the question is less well resolved, and other determination methods may be preferable.

REFERENCES

(2) HAXEL, O. and HOUTERMANS, F. G., Z. Physik. 124, 705 (1948).
II. 2. 1. 1. DEMOUNTABLE 4π GEIGER COUNTERS FOR
STANDARDIZATION AND THE ASSAY OF LOW-
ACTIVITY SAMPLES

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Abstract

Some developments in the design of 4 π GM counters are reported and
applications of these counters in the measurements of radioactive standards, in
relation to radiological work, are discussed. In the two types of 4 π GM
counter described the anode wires can be easily removed thereby facilitating
cleaning.

One type has been used successfully for several years in the standardization
of β-emitting radioactive nuclides and some results of intercomparisons of
standards with other laboratories are presented. These counters have also been
applied, in conjunction with a γ ionization system, in determinations of the
specific γ-ray emission (k-factor) for Fe⁵⁹, Cs¹³⁷ and Ir¹⁹².

The second type is a smaller low-background version of the above counter,
constructed in stainless steel and designed primarily for the assay of small
amounts of radioactivity in biological materials. It can also be adapted for the
absolute measurement of radioactive sources at low counting rates (≤ 100
counts per minute) since, when shielded with cosmic-ray counters in anti-
coincidence, a stable background of between 1 and 2 counts per minute can
be obtained.

Compteurs 4 π GM démontables pour l'étalonnage et l'essai d'échantillons de
faibleactivité

Les auteurs décrivent certains perfectionnements dans la construction de
compteurs 4 π GM et discutent les utilisations de ces compteurs pour la mesure
d'étalons radioactifs, en relation avec les recherches radiologiques. Les filaments
anodiques des deux types de compteurs 4 π GM décrits peuvent être facilement
enlevés, ce qui facilite le nettoyage.

L'un des types a été utilisé avec succès durant plusieurs années pour
l'étalonnage de nuclides β actifs; les auteurs présentent les résultats des com-
paraisons d'étalons faites avec d'autres laboratoires. Ce type de compteur a été
egalement utilisé pour la détermination du facteur k (émission γ spécifique) de
Fe⁵⁹, Cs¹³⁷ et Ir¹⁹².

Le second type de compteurs est une version réduite, à faible mouvement
propre, du premier. Construit en acier inoxydable, il a été initialement conçu
pour l'essai de faibles activités dans les matériaux biologiques. Il peut cependant
être adapté à la mesure absolue de sources de faible niveau d'activité (≤ 100 cpm)
car, avec une gaine de compteurs cosmiques en anticoincidence son mouvement
propre s'abaisse jusqu'à 1—2 cpm.
«Съемные счетчики Гейгера на 4 л для стандартизации и оценки проб низкой активности»

Излагаются некоторые результаты работы по конструированию счетчиков Гейгера-Мюллера на 4 л, а также вопрос их использования для измерения стандартов радиоактивности в работе с радиоактивными материалами. В двух видах счетчиков на 4 л, которые рассмотрены в данном документе, анодные провода легко снимаются, благодаря чему облегчается чистка.

Один из счетчиков успешно использовался в течение ряда лет для стандартизации β-излучающих радиоактивных нуклидов, и в документе приводятся некоторые результаты сопоставления стандартов со стандартами, полученными другими лабораториями. Этот счетчик использовался также вместе с γ ионизирующей системой для определения γ излучений Fe⁵⁹, Cs¹³⁷ и Ir¹⁹².

Другой счетчик является счетчиком низкого фона. Он устроен так же как и вышеуказанный счетчик, изготовлен из нержавеющей стали и предназначен главным образом для оценки небольших количеств радиоактивности в органических веществах. Он может также быть приспособлен для абсолютного измерения радиоактивных источников на малых скоростях счета (≤100 отсчетов в минуту), поскольку можно получить стабильный фон (1—2 счета в минуту), если одновременно для счёт несовпадений использовать счетчики космических лучей.

Condatores GM 4 π para la normalización de patrones radiactivos con baja actividad

Los autores dan cuenta de algunos perfeccionamientos introducidos en la construcción de contadores Geiger-Müller 4 π y estudian las aplicaciones de estos aparatos en la medición de patrones radiactivos utilizados en trabajos de radiología. En los dos modelos de contadores Geiger-Müller 4 π que los autores describen los alambres anódicos pueden desmontarse fácilmente, lo cual facilita su limpieza.

Uno de los modelos de contadores se viene utilizando con éxito desde hace varios años para normalizar radionúclidos β; se exponen algunos resultados de las comparaciones efectuadas con los patrones de otros laboratorios. Estos contadores también se han empleado, en combinación con un dispositivo de ionización γ, para determinar la emisión γ específica (factor k) del Fe⁵⁹, Cs¹³⁷ y Ir¹⁹².

El segundo modelo, de acero inoxidable, es una copia del primero, en menor escala; está diseñado para trabajar con bajas actividades ambiente y se destina principalmente a la determinación cuantitativa de pequeñas cantidades de substancias radiactivas contenidas en materiales biológicos. También puede adaptarse para la medición absoluta de fuentes radiactivas de bajo índice de recuento (≤ 100 impulsos por minuto), pues si se le rodea de contadores de rayos cósmicos, montados en anticoincidencia, es posible estabilizar la actividad de fondo en 1 a 2 impulsos por minuto.
Introduction

We have found it valuable to have readily available in this laboratory methods for preparing standards, by absolute measurements, in relation to the therapeutic and diagnostic applications of radioactive nuclides. We have paid most attention to the use of $4\pi$ counting techniques, including liquid scintillation (1) and GM counters (2), and also to $\gamma$ ionization measurement with cavity chambers (4). In addition, we have been increasingly concerned with problems of environmental radioactivity, both fission products (3) and naturally occurring radionuclides, and this has led us to attempt to use $4\pi$ counters in anti-coincidence counting assemblies, both for assay and standardization.

Demountable $4\pi$ GM counter for normal laboratory use

The constructional details of the $4\pi$ GM counter that we have developed for use in absolute measurements of $\beta$-emitting nuclides are shown in figure 1 and Table I. The anode wires are mounted on a framework which can be
TABLE I
Constructional Details of 4π Counters

<table>
<thead>
<tr>
<th>Counter</th>
<th>Cathode &amp; anode framework</th>
<th>Insulators</th>
<th>Anode</th>
<th>Active length cm</th>
<th>Internal diameter cm</th>
<th>Anode wire diameter mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Large</td>
<td>Brass</td>
<td>Glass</td>
<td>Tungsten</td>
<td>12</td>
<td>3.6</td>
<td>0.1</td>
</tr>
<tr>
<td>Small</td>
<td>Stainless steel</td>
<td>Polytetrafluoroethylene (PTFE) &amp; ceramic</td>
<td>Tungsten</td>
<td>2.5</td>
<td>2.5</td>
<td>0.1</td>
</tr>
</tbody>
</table>

readily removed from the cathode, vacuum seals being formed by 0-rings at each end. This method of construction, which was introduced to simplify assembly, inspection of the internal surfaces and cleaning, has proved very satisfactory (2).

Two counters of this type have been used frequently over the past four years for the intercomparison of standards with members of the National Physical Laboratory Advisory Committee on Radioactive Standards and results obtained are summarized in Table II. The discrepancy between our values and those obtained by NPL has been consistently $\pm 3\%$, except for the samples of Tl$^{204}$ and Sr$^{85}$.

To illustrate the applications of these counters we shall discuss our investigations of Cs$^{137}$, a fission product which is finding important uses in radiotherapy and is also a readily observable contaminant in the environment (3).

TABLE II
Intercomparison of Standard Measurements 1955—1958

<table>
<thead>
<tr>
<th>Nuclide (Chronological order of measurement)</th>
<th>Institute of Cancer Research</th>
<th>NPL</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tl$^{204}$</td>
<td>1.04</td>
<td></td>
</tr>
<tr>
<td>Sr$^{85}$</td>
<td>0.96</td>
<td></td>
</tr>
<tr>
<td>Na$^{24}$</td>
<td>1.03, 1.00, 1.00, 0.98 **</td>
<td></td>
</tr>
<tr>
<td>Fe$^{59}$</td>
<td>1.02</td>
<td></td>
</tr>
<tr>
<td>Ca$^{45}$</td>
<td>0.98</td>
<td></td>
</tr>
<tr>
<td>Y$^{90}$</td>
<td>1.01</td>
<td></td>
</tr>
<tr>
<td>I$^{131}$</td>
<td>1.02, 0.97 **</td>
<td></td>
</tr>
<tr>
<td>Se$^{80}$ + Y$^{90}$</td>
<td>0.99</td>
<td></td>
</tr>
<tr>
<td>Au$^{198}$</td>
<td>0.99</td>
<td></td>
</tr>
</tbody>
</table>

$^a$ R = ratio of specific activities, referred to agreed reference time, derived from measurements at the two laboratories of sources prepared independently from a stock solution. Measurements were made at approximately the same time at each laboratory. At NPL a 4πβ proportional counter was used throughout this work.

$^{**}$ Results for measurements of several standards.
It therefore became necessary for us to prepare standards of this nuclide and a
determination of the specific \( \gamma \)-ray emission (k-factor) for Cs\(^{137} \) + Ba\(^{137m} \) was
made \(^2\) by combining \( 4 \pi \beta \) counting of suitable sources with measurements of exposure dose rate using the \( \gamma \) ionization system referred to above.

The Cs\(^{137} \) sample contained a low but significant proportion of Cs\(^{134} \), which
was estimated by scintillation spectrometry to be 1% of the activity due to Cs\(^{137} \).

The measured \( 4 \pi \beta \) counting rates were corrected for this Cs\(^{134} \) conta-
mination and also for the Ba\(^{137m} \) internal conversion electrons (using two alter-
native decay schemes discussed below). The \( \gamma \) ionization measurements were also
corrected for Cs\(^{134} \) contamination using a theoretical k-factor of 9.3 r cm\(^2\) mc\(^{-1}\) hr\(^{-1}\). It was estimated that the contribution by K-x-rays from Ba to the
measured exposure dose rate for the source used did not exceed 1%.

The theoretical k-factor derived from the decay scheme of Ricci \(^4\) is
3.25 r cm\(^2\) mc\(^{-1}\) hr\(^{-1}\) (96% transitions to Ba\(^{137m} \), \( e/\gamma = 0.114 \)). The k-factor
derived from our experimental measurements is 3.46 r cm\(^2\) mc\(^{-1}\) hr\(^{-1}\). The ratio
between these values is 1.06 whilst the estimated experimental error is \( \pm 5\% \).

On the other hand, for the widely used decay scheme set out by STROMINGER
et al. \(^5\) (92% transitions to Ba\(^{137m} \), assumed \( e/\gamma = 0.114 \)) the theoretical and
corrected experimental values are 3.11 and 3.45 r cm\(^2\) mc\(^{-1}\) hr\(^{-1}\) respectively,
the discrepancy between them exceeding the estimated limits of error in our
measurements.

Low-Background \( 4 \pi \) GM counters

A method of assessing the relative merits of counters with different
efficiencies and backgrounds has been given by LOEVINGER et al. \(^6\). Equation
(15) of their paper can be reduced to the form
\[
F = \frac{100}{E \sqrt{T}} = \frac{S}{\sqrt{m(S+B)} + \sqrt{B}}
\]
where \( F \) is the "factor of merit" of the counter
\( m = \) number of samples counted for each measurement of background.
\( S = \) average counting rate above background of the \( m \) samples (assuming
roughly equal counting rates for each sample).
\( B = \) background counting rate.
\( T = \) total time spent in counting the background and \( m \) samples.
\( E = \) Standard deviation as \( \% \) of \( S \).

For \( S < B \), equation (i) approximates to the well-known result:
\[
\frac{1}{E \sqrt{T}} \propto \frac{S}{\sqrt{B}}
\]
It should be noted however that this approximation is frequently applied in
circumstances where the full expression should be used.

It follows from equation (i) that it is worth while increasing the efficiency
of a detector in order to obtain a higher value of \( F \) \(^7\). Any changes made will
probably also raise the value of \( B \) but an increase in \( F \) may still result. For
example if both \( S \) \& \( B \) are doubled the value of \( F \) is increased by \( \sqrt{2} \) and the
time required for measurement with the same error is halved.

As the highest efficiency can be obtained using a \( 4 \pi \) counter, we designed
a low-background version of such a counter for the assay of low-activity
sources derived from various biological materials (8). Limitations imposed by chemical procedures precluded the use of samples greater than 50 mg in weight, many of which were too weak to be measured on our routine low-background end-window counters (5). During development it became apparent that the instrument would also be suitable for making absolute measurements at counting rates of about 1 cps (to be compared with the values of 100—200 cps normally used) on sources prepared from solutions with specific activities as small as $2.5 \times 10^{-3} \mu\text{c/g}$. Standard solutions of such a strength are required for the calibration of our routine low-background counters. These solutions may be prepared by diluting a stronger solution standardized in the larger $4\pi$ counter. However, it is necessary to check the accuracy of dilutions, and we have found that this is best done by making absolute measurements on the weak solutions using the low-background $4\pi$ counters. A further advantage of this procedure is that contamination introduced by reagents can be readily detected, since the sensitivity of the $4\pi$ counters for thin sources is almost independent of $\beta$ particle energy.

![Figure 2](attachment:image.png)

*Anode structure of demountable low-background $4\pi$ GM counter for low-level standardization and assay.*

The design was based on the $4\pi$ standards counter described above but with care taken to minimize the background by reducing the total size and by using materials expected to be low in radioactive content (9). Constructional details are given in figure 2 and Table I. The slide is so made that a thick source mounted between sheets of aluminized Melinex can be inserted. The plateaux obtained are more than 150 volts long with slopes of about 3% per 100 volts.

In developing this counter and testing its performance three main features had to be considered:

(i) Efficiency

The efficiency for point sources of $\text{Sr}^{90} + \text{Y}^{90}$ was examined and found to be identical, within experimental error, with that obtained using the larger
standards counter at the same filling pressure (20 cm Hg argon, 1 cm Hg ethyl formate).

As an illustration of the efficiency obtainable using thick sources (50 mg/cm²) of KCl we may cite comparative K⁴⁰ measurements on the low-background 4π counter and an end-window counter (Gen. Elect. Co. type GM 4 LB, having a 7 mg/cm² duralumin window). The efficiencies based on chemical estimation of the potassium are 70% and 20% respectively.

(ii) Background

These small 4π counters have background of approximately 1 cpm when operated in anti-coincidence with an assembly of cosmic-ray counters, shielded by 2.5 cm of aged lead and placed 2 metres deep (figure 3) in chalk (radioactive

<table>
<thead>
<tr>
<th>Counter</th>
<th>Shielding</th>
<th>Source</th>
<th>B cpm</th>
<th>S cpm</th>
<th>$S/\sqrt{B}$</th>
<th>F for m = 5</th>
<th>T min (for S. D. ± 3%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Large</td>
<td>5 cm Pb</td>
<td>25 μμ C</td>
<td>55</td>
<td>100</td>
<td>5.5</td>
<td>1.5</td>
<td>530</td>
</tr>
<tr>
<td>Small</td>
<td>2 m chalk + 2.5 cm Pb + anti-coincidence shield.</td>
<td>25 μ C</td>
<td>55</td>
<td>1.3</td>
<td>48</td>
<td>3.6</td>
<td>100</td>
</tr>
</tbody>
</table>

TABLE III

Comparison of factor of merit for 4π counters

![Figure 3](image_url)

Pit, excavated in chalk with galleries cut in the side walls, used for low-background measurements (reproduced by permission of the Editor, Nature).
content 7 to $8 \times 10^{-13}$ curie $\alpha$ activity/g) (10). The background for one of the large $4\pi$ GM counters was 20 cpm under the same conditions.

The total counting time $T$ required to assay a sample of 25 $\mu$Ci using the two types of counter is shown in Table III, in which it is assumed that 5 points on the plateau are to be measured, each with an accuracy of $\pm 3\%$. In addition the factor of merit $F$, calculated using equation (i), is given. It should be noted that, using this equation, the relative increase in $F$ is 2.5 whereas using the approximate expression $F \propto \frac{S}{\sqrt{B}}$ the relative increase is 8.7.

(iii) Stability

For the measurement of weak sources obtained from environmental material, these counters (unlike those used in normal standards work) have to be operated with single fillings for periods ranging from a few hours to a day or more. Precautions had therefore to be taken to minimize the passage of oxygen or water vapour into the counters.

It is well known that traces of air can cause an increase in plateau slope (11); we have found a significant effect for pressures of air exceeding 20 $\mu$. Accordingly, after evacuating the counter with a diffusion pump to 0.03 $\mu$, a check is made using a Penning gauge that the pressure has not risen by more than 0.15 $\mu$ 10 minutes after the counter has been isolated from the pump. We may then expect to be able to operate the counter for about 1000 minutes before any leak becomes significant.

![Figure 4](image.png)

Figure 4

Stability of counter for repeated refilling, (i) with, and (ii) without, anti-coincidence cosmic-ray cancellation. Points A and B are total counting rates ($S+B$) for samples, all other points background counting rate only. All errors shown are $\pm \sigma$. 

182
In order to reduce the possibility of desorption of gases by the O-rings they are made from a compound (AM 55, Dowty Seals Ltd.) which the manufacturers claim does not desorb gases.

Using 1 cm ethyl formate (SVP 20 cm Hg) and 20 cm Hg argon as filling mixture, we have observed a slow decrease in starting potential after filling, sometimes by as much as 100 volts in 24 hours. However, using methyl formate (SVP 50 cm Hg) as quenching agent, only a slight change of 10—20 volts is observed. This improvement is probably a result of reduced adsorption of the more volatile compound.

Figure 4 (i) shows the reproducibility that can be obtained in the measurement of background and of very weak samples (A and B) when the counter is operated in the anti-coincidence shielded system described above. Each point represents approximately 1000 minutes counting time. Sample B is at the limit of practicable measurement (0.1 cpm) with an estimated error of ±0.05 cpm. The importance of using anti-coincidence in making such tests is illustrated by the marked fluctuations of background shown in figure 4 (ii).

In conclusion, we have shown that, taking the precautions discussed, a readily demountable 4 \( \pi \) GM counter can be used successfully to measure very weak samples over long counting times. Measurement for about twenty hours is our normal practice, but it has been found possible on occasion to operate on single fillings for as long as three days.

Acknowledgements

The encouragement of Professor W. V. Mayneord throughout this work is gratefully acknowledged. We should like to express our appreciation of the co-operation we always received from the late Mr. W. E. Perry, National Physical Laboratory and to thank Dr. B. W. Robinson for permission to present the results of intercomparison of standards with his Division at that laboratory. We would like to thank Mr. R. H. Parkes of Accles and Pollock Ltd for the samples of stainless steel used for the low-background counters.

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REFERENCES


II. 2. 2. SELF-ABSORPTION STUDIES WITH A 4π $\beta$-PROPORTIONAL FLOW COUNTER

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Self-absorption studies of $\beta$ emitters have been made by means of the techniques outlined by Pate and Yaffe [Can. J. Chem. 34, 265 (1956)]. Volatile compounds of H$_3$, C$_{14}$, Co$^{60}$, Hg$^{203}$, As$^{77}$, and P$_{32}$ ($E_{\text{max}}$ ranging from 0.018 to 1.7 MeV) were distilled on to thin VYNS mounts and the self-absorption characteristics determined. An empirically determined series of curves relating $E_{\text{max}}$, source thickness, and self-absorption is given.

The self-absorption data have been subjected to a theoretical analysis for counters of small angle and $4\pi$ geometry. The experimentally determined self-absorption curves for $4\pi$ geometry lie in between the theoretical small angles and $4\pi$ geometry curves.
Estudios sobre autoabsorción con un contador de flujo proporcional $\beta 4 \pi$

Les autores han estudiado la autoabsorción de los emisores $\beta$ utilizando las técnicas descritas por Pate y Yaffe [Can. J. Chem. 34, 265 (1956)]. Han destilado compuestos volátiles de $\text{H}_3$, $\text{C}^{14}$, $\text{Co}^{60}$, $\text{Hg}^{203}$, $\text{As}^{77}$ y $\text{P}^{32}$ ($E_{\text{max}}$ comprendida entre 0,018 y 1,7 MeV) recogiendo los sobre finas películas de resina vinílica (VYNS) y han determinado las características de autoabsorción. Presentan una serie de curvas, obtenidas experimentalmente, que relacionan $E_{\text{max}}$ el espesor de la fuente y la autoabsorción.

Les datos referentes a la autoabsorción han sido sometidos a un análisis teórico, para contadores de ángulo pequeño y $4 \pi$. Las curvas de autoabsorción determinadas experimentalmente para contadores $4 \pi$ están comprendidas entre las curvas teóricas que corresponden a los ángulos sólidos pequeños y las curvas determinadas mediante contador de geometría $4 \pi$.

Dr. Pate in his review paper (II. 2. 1) has described for you the application of the $4 \pi \beta$ counter to the measurement of the absolute disintegration rates of radioactive substances. He has pointed out the variables which have to be considered in order to obtain an absolute disintegration rate and told you of the progress which has been made with these. In particular, he has indicated that the problem of self-absorption still remains.

Very little work has been done on this aspect of this field. SELIGER and SCHWEBEL (1) have emphasized the difficulties encountered in measuring $\text{Sr}^{90}$ and $\text{Co}^{60}$ sources prepared in different ways. MEYER-SCHUTZMEISTER and VINCENT (2) obtained differences of the order of $30\%$ in counting rate by depositing $\text{S}^{35}$ in crystals of varying diameter.

HAWKINGS et al. (3) proposed the method of progressive dilution until a constant specific activity (after correction for the dilution) was obtained. Alternatively, the logarithm of the counting rate could be plotted against dilution and the curve extrapolated to infinite dilution. This method, however, while certainly an improvement over existing techniques where self-absorption is ignored, is hardly the final answer. As pointed out (4) the material still crystallizes into local aggregates and reduction of crystal size occurs only up to a certain point, beyond which dilution is of very little help, other than to separate the aggregates.

MERRITT et al. (5) in a paper published recently and in the paper which Dr. Campion will be presenting in this session (II. 2. 4) have adopted several new and significant approaches to this problem. I am grateful to them for allowing me to see the preprint of their paper but will refrain from discussing it since it will be presented shortly.

In this paper I should like to discuss some work which has been done in our laboratory in this field and to describe attempts which are being made to clear up this last remaining difficulty.

Dr. Pate has described some of the problems concerned with source preparation and has shown you the results obtained by workers using various methods of source preparation. In particular he has rather briefly described
Figure 1
Distillation Apparatus.

Figure 2
Uniformity of distilled sources.
the method of vacuum distillation, and I would like to continue this dis-
cussion. The apparatus which we have employed is essentially the same as that
described by PATE and YAFFE (4) and is shown in figure 1. It consists of a
platinum micro-crucible, A, which is lowered into position in a Pyrex chamber
connected to a vacuum system. Over the top of the crucible is placed a collimat-
ing diaphragm which has a circular opening 1 cm$^2$ in area. The crucible is
supported by an aluminium ring, E, which is thermally insulated from the
crucible by a mica ring, and is heated by means of the work-coil, B, of a radio-
frequency generator. The temperature can easily be measured by a thermo-
couple arrangement, D. Into the crucible is placed the active material which I
shall describe shortly. The crucible can be heated easily to temperatures of the
order of 1000° C. The active material distils on to a very thin VYNS film, F,
(of the order of 5 μg/cm$^2$), and the film shows no deterioration at these high
temperatures. If quantitative transfer is desired, this can be accomplished by
removing the collimating ring and placing the film very close to the mouth
of the crucible. One can with very little practice obtain an exceedingly
uniform deposit, and in figure 2 is shown some measurements of the uniformity.

Although one can very easily distil inorganic compounds, the best results
we have obtained to date are with compounds containing organic groupings.
These compounds are readily prepared, incorporating a great variety of radio-
active species. Volatilization with the compounds used was satisfactory at
the pressure range employed (of the order of 1 μ Hg) and at moderate crucible
temperatures. Temperatures can be controlled rather easily to ± 10° C, and
the slower the rate of distillation the more uniform the deposit. Electron
micrograph studies showed that the crystals were uniformly small, of the order
of 1 μ diameter.

Spectrophotometric determinations, taken before and after distillation,
showed that there was no destruction of the solid compound which was used.
In this manner the following nuclides were volatilized on to films: H$^3$, Ni$^{63}$,
C$^{14}$, Hg$^{203}$, Co$^{60}$; As$^{77}$ and P$^{32}$. This gave a wide range of β− emitters with
maximum energies varying from 0.018 to 1.7 MeV. The organic compounds
used were prepared by variants of standard techniques and are shown in
Table I.

**TABLE I**

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>$E_{max}$ (MeV)</th>
<th>Compound</th>
<th>Distillation Temp. (°C)</th>
<th>Pressure (μ Hg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>H$^3$</td>
<td></td>
<td>Benzidine</td>
<td>220</td>
<td>1</td>
</tr>
<tr>
<td>Ni$^{63}$</td>
<td></td>
<td>Nickel dimethylglyoxime</td>
<td>120</td>
<td>1</td>
</tr>
<tr>
<td>C$^{14}$</td>
<td></td>
<td>Methionine</td>
<td>250</td>
<td>1</td>
</tr>
<tr>
<td>Hg$^{203}$</td>
<td></td>
<td>Aminomericuric chloride</td>
<td>360</td>
<td>1</td>
</tr>
<tr>
<td>Co$^{60}$</td>
<td></td>
<td>Cobalt α-nitroso-β-naphthalate</td>
<td>270</td>
<td>1</td>
</tr>
<tr>
<td>As$^{77}$</td>
<td></td>
<td>Phenarsazine</td>
<td>200</td>
<td>1</td>
</tr>
<tr>
<td>P$^{32}$</td>
<td></td>
<td>Benzidine phosphate</td>
<td>235</td>
<td>5</td>
</tr>
</tbody>
</table>
The data on Ni$^{63}$, obtained by using nickel dimethylglyoxime are those previously described by PATE and YAFFE (4) and are included for completeness.

The mass thicknesses were determined with a Beckman DU spectrophotometer. After activity measurements had been made, the active sources were dissolved off the film, the film checked in a counter to ensure that the source had been completely removed, and the amount of material determined by measuring the optical transmission at appropriate wave-lengths.

The self-absorption data were obtained using a counter of the type described by Dr. Pate, and this is shown in figure 3 to refresh your memory. The self-absorption data are shown in figures 4—10 inclusive. Here we plot the apparent relative specific activity against the superficial density of the source.
Figure 4
Self-absorption curves — H^3

Figure 5
Self-absorption curves — Ni^{63}

Figure 6
Self-absorption curves — C^{14}

Figure 7
Self-absorption curves — Hg^{203}
In Figure 11 is given a cumulative curve where the apparent relative specific activity is plotted against $\beta$ end-point energy. This gives a curve which is very useful in the practical sense. It can be used for determining the self-absorption of sources prepared in the manner described and indeed, if one prepares, in some other manner, sources which are as uniformly distributed as these are, then one also ought to be able to use these curves.

A word of caution is in order here. One must realize that if the spectral shape of a $\beta$ emitter differs radically from that of the $\beta$ emitter which has been used in this investigation, then the use of these curves will represent only a first approximation. However, you can see that in most cases the error will be slight.

It is of interest to determine something about the manner in which self-absorption can be described theoretically under these conditions. A great deal of work has been done in this respect dealing with self-absorption in the conventional small-angle geometry systems. In the main, however, the relationships developed are empirical and do not take any account of the geometry of the systems used.

The simplest of these is based on the assumption that the absorption of $\beta$ radiation resulted in an exponential decrease of radiation intensity with increasing sample thickness, so that

$$a = \frac{a_0}{\mu d} \left(1 - e^{-\mu d}\right) \quad \ldots \ldots \ldots \quad (1)$$
Figure 10
Self-absorption curves — P₃²

Figure 11
Cumulative self-absorption curves.

BENZIDINE PHOSPHATE

ENERGY (MEV)

SUPERFICIAL DENSITY OF SOURCE (μG/M²)

SELF-ABSORPTION FACTOR

ENERGY (MEV)
Here $a$ is the observed specific activity of a sample of specific activity $a_0$, $d$ is the superficial density of the sample and $\mu$ the absorption coefficient.

ELLIOTT and co-workers \cite{6} attempted to bring in the angle of observation of the radiation, i.e. the angle subtended by the counter at the source. If the observation is made at an angle $\Theta$ to the normal, then the effective source thickness becomes \[ \frac{d}{\cos \Theta} \text{, and} \]

\[ a_{\phi} = \frac{a_0 \cos \Theta}{\mu d} \left(1 - e^{-\mu d / \cos \Theta}\right) \] . . . . . . (2)

They showed that the radiation from sources of finite thickness was anisotropic, the radiation being concentrated more and more in the direction of the normal to the source plane. This meant that self-absorption effects depended on the angle of observation and would be minimal for a small geometry counting system as exemplified by the typical end-window counter.

REID et al. \cite{7} evolved a theoretical expression to describe the dependence of self-absorption on counter geometry. If one integrates equation (2) over a cone of half-angle $\alpha$ (representing the solid angle over which radiation from a point source is detected) the specific activity $a_{\alpha, d}$, observed under these conditions for source thickness $d$, is

\[ a_{\alpha, d} = 2\pi \int_{0}^{\alpha} a_{\theta, d} \sin \Theta \, d\Theta \]

where $2\pi \sin \Theta \, d\Theta$ is the solid angle element of integration. If one substitutes for $a_{\theta, d}$ and replaces $a_0$ by $a_0$, the specific activity per unit angle, the above expression on expansion will give

\[ \frac{a_{\alpha, d}}{a_0} = \frac{1}{2} \left[ e^{-\mu d} - \frac{\mu^2 d^2}{2!} + \frac{\mu^3 d^3}{3!} - \frac{\mu^4 d^4}{4!} + \ldots \right] \]

\[ - \frac{1}{2} \left[ \cos \alpha e^{-\mu d / \cos \alpha} - \mu d \ln \cos \alpha - \frac{\mu^2 d^2}{\cos \alpha} + \ldots \right] \ldots (3) \]

The above expression was checked by YANKWICH et al. \cite{8} for $^{14}$C and $^{35}$S in geometrical arrangements varying from 7.6 to 20.5% of $2 \pi$ steradians.

A more rigorous check on the effect of geometry was made by BAKER and KATZ \cite{9} who used an array of Geiger-Mueller tubes arranged around the sample to simulate a $4 \pi$ geometrical arrangement. They found the effects for thick samples to be described adequately by the simple exponential self-absorption formula. The following relation

\[ \mu = 0.0155 \, E_m^{-1.41}, \] . . . . . . . . (4)

where $E_m$ the maximum energy of the $\beta$ emitter is in MeV and $\mu$, the absorption coefficient is in cm$^2$/mg, was determined to be applicable. The expression was used by the authors for $\beta$ emitters with $E_m$ ranging from 0 to 3 MeV and values of $d$ from 0—500 mg/cm$^2$. 

Baker and Katz further showed that

\[ f_{\alpha} = \frac{1}{(1 - \cos \alpha)} \frac{1 - e^{-\mu d}}{\mu d} \int_{0}^{\alpha} \frac{\alpha \, \theta, d}{a_{d}} \sin \Theta \, d\Theta \ldots \ldots \ldots (5) \]
Where $f_\alpha$ = self-absorption factor
and $a_d$ = number of $\beta$ particles leaving the surfaces of the source.

As $\Theta \to 0$, this equation reduces to

$$f_0 = \frac{a_{a,d}}{a_d} \cdot \frac{1 - e^{-\mu d}}{\mu d} \quad \ldots \ldots \ldots \ldots \ldots (6)$$

and

$$f_{4\pi} = \frac{1 - e^{-\mu d}}{\mu d} \quad \ldots \ldots \ldots \ldots \ldots (7)$$

The authors point out, however, that the factor $\frac{a_{a,d}}{a_d}$ has a value ot 2 for $d$ values in excess of 0.2 R. If one extrapolates the curve obtained from equation 7 to a zero value of $d$, this will lead to a corrected apparent disintegration rate twice the true value.

A rigorous integration procedure is given by GORA and HICKEY (10). Their starting integral

$$a_{a,d} = \frac{1}{\Omega_a} \int_0^\alpha a_0 \cos \Theta \frac{d}{d\Omega} \left(1 - e^{-\mu d/cos \Theta}\right) d\Omega \quad \ldots \ldots \ldots \ldots (8)$$

is analogous to that of REID et al. (7), except that the volume element is written as

$$d\Omega = 2\pi \sin \Theta \ d\Theta$$

and the expression is divided through by

$$\Omega = 2\pi (1 - \cos \alpha),$$

so that $a_{a,d}$ becomes $a_\alpha$, the observed counting rate in a cone of half angle $\alpha$ when $d = 0$, i.e. for an infinitely thin source. If $\lambda$ is written for $\mu d$ and

$$y = \frac{\lambda}{\cos \Theta},$$

then equation (8) becomes

$$a_{a,\lambda} = \frac{a_0 \lambda}{1 - \cos \alpha} \int_0^{\lambda/cos \alpha} \left(1 - e^{-\lambda y/y^3}\right) dy$$

$$= a_0 f(\alpha, \lambda)$$

where

$$f(\alpha, \lambda) = \left[F(\lambda) - \cos \alpha F\left(\frac{\lambda}{\cos \alpha}\right)\right]/(1 - \cos \alpha)$$

and

$$F(\lambda) = \frac{1}{2} \left[1 - e^{-\frac{\lambda}{\lambda}} + e^{-\lambda} + \lambda E_1(-\lambda)\right].$$

The exponential integral function $E_1(-\lambda) = -\int_{-\lambda}^\infty \frac{e^{-t}}{t} dt$ has been tabulated (11).
The limiting cases of this result are as follows:

(a) \( \lambda \to 0 \), i.e. small \( \mu \) or \( d \), then

\[
F(\lambda) \sim 1, \quad f(\alpha, \lambda) \sim 1, \quad \text{and} \quad a_{\alpha, \lambda} = a_0
\]

i.e. no absorption effects.

(b) \( \lambda \gg 1 \), i.e. \( d \) large, then

\[
F(\lambda) \sim \frac{1}{2\lambda},
\]

and

\[
f(\alpha, \lambda) = \frac{1}{2\lambda} - \left( \frac{\cos^2 \alpha}{2\lambda} \right)
\]

\[
= \frac{1 + \cos \alpha}{2\lambda}
\]

i.e.

\[
f(\alpha, \lambda) = \frac{1}{\lambda}
\]

and

\[
f\left(\frac{\pi}{2}, \lambda\right) = \frac{1}{2\lambda}.
\]

Thus the observed specific activity \( a_0 \), \( \lambda \) should decrease about twice as quickly with increasing \( d \) in a 2\( \pi \) steradian geometrical counting assembly as in a narrow angle one. GORA and HICKEY further point out that it is meaningless to quote absorption coefficients unless the geometrical efficiency, for which this is measured, is obtained. These authors provide data obtained with 2\( \pi \) steradians and narrow angle geometries, and good agreement is observed. This is despite the fact that (a) the usual approximation of exponential absorption is used and (b) no account is taken of the self-scattering processes in the source, unless the cosine \( \Theta \) radiation distribution takes this into account.

We have fitted our data to the Gora and Hickey equation which they derived for a 2\( \pi \) steradian geometry, and this is identical for a 4\( \pi \) steradian geometry. With the exception of \( \text{H}^3 \) and \( \text{C}^{14} \), our experimental results gave a very good fit. The absorption coefficient \( \mu \) was derived in this manner using data up to a superficial density of 100 \( \mu g/cm^2 \), and the results are plotted in figure 12. One notes that the value for \( \text{C}^{14} \) is discordant. This may be due to the large number of soft electrons observed in the spectrum. In Table II are compared the values of absorption coefficients which one obtains using the Gora-Hickey and Baker-Katz formalisms, along with that which would be obtained if one assumed exponential absorption only.

That the latter assumption is a gross approximation was of course evident from the nature of the absorption curves of the softer \( \beta \) emitters, and it is indeed probable that the same behaviour would be observed with the more energetic \( \beta \) emitters at greater superficial densities. The difference between our values and those of Baker and Katz is hardly surprising since they used much thicker samples and their absorption coefficients are probably limiting values.

The absorption coefficient then appears to be a very complex function, probably not constant, but continuously variable depending on sample thickness, the spectrum of the nuclide, and the geometry of the counting arrangement.
The scattering in the sample itself is what makes the exponential assumption even possible.

Because of these considerations, it is exceedingly difficult for any mathematical comparison of the results with theory to be other than an approximation. To do this we have used the Glendenin range-energy relationship (12)
Figure 14  
Self-absorption curve for $\text{S}^{35}$ — experimental and theoretical.

Figure 15  
Self-absorption curve for $\text{P}^{32}$ — experimental and theoretical.

Figure 16  
Self-absorption curve for $\text{H}^8$ — experimental and theoretical.

Figure 17  
Self-absorption curve for $\text{C}^{14}$ — experimental and theoretical.
extended to lower energies by the addition of data from RUTHERFORD, CHADWICK and ELLIS (13). To obtain absorption curves in this fashion one needs to transform the spectral data into a Kurie plot and extrapolate to zero energies. This was done using Fermi β decay theory with the Konopinski-Uhlenbeck corrections for forbidden transitions.

Each spectrum was split into 18 parts in this manner and the absorption curve obtained. The curves so obtained are those expected in a narrow angle geometry, since no allowance for oblique paths through the absorber has been made. For radiation emitted into a solid angle of 2 \( \pi \) steradians, it is necessary to allow for increased absorption for those particles having a path length greater than \( d \). This is done in a manner analogous to GORA and HICKEY (10).

As can be seen from figures 13—17 inclusive, the experimental 4 \( \pi \) results fall between the predicted small angle and predicted 4 \( \pi \) absorption curves. Better agreement is hardly to be expected since absorption coefficients applicable to only the first 100 \( \mu \mathrm{g/cm}^2 \) superficial density were used. The agreement in this area is qualitatively good and shows the need for increasing the complexity of the system by incorporating either a series of absorption coefficients or a continuously varying one in the calculations.
REFERENCES


II. 2. 3. ON ABSOLUTE MEASUREMENTS OF \( \beta \)-EMITTING RADIONUCLIDES

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Abstract

\( 4 \pi \) GM detectors are described and some of their characteristics presented.

The absence of spurious pulses is shown by the measurement of the rate of arrival of intervals between pulses.

Some problems related to the foil absorption and source absorption are considered using \( \text{Sr}^{90} \) and \( \text{Y}^{90} \).

Aiming at correcting the counting losses in the foil and in a certain extent also the source absorption an experimental method which uses paired sources is presented.

The preparation of standards of \( \text{Bi}^{210} \) (RaE) is described.

A note is presented on the problem of foil absorption losses in scintillation absolute counting.

Mesures absolues d'émetteurs \( \beta \)

Le mémoire décrit des détecteurs \( 4 \pi \) GM et donne certaines de leurs caractéristiques.

L'absence d'impulsions parasites est indiquée par la mesure de la cadence d'arrivée des impulsions.

L'auteur examine plusieurs problèmes que posent l'absorption par la source et l'absorption par la feuille lorsqu'on utilise du \( \text{Sr}^{90} \) et du \( \text{Y}^{90} \).

Il expose une méthode expérimentale dans laquelle il emploie des sources deux à deux en vue de corriger les pertes au comptage dans la feuille et aussi, dans une certaine mesure, l'absorption par la source.

L'auteur décrit la préparation d'étalon de \( \text{Bi}^{210} \) (RaE).

Il ajoute une note sur le problème des pertes par absorption dans la feuille dans le comptage absolu par scintillations.

Абсолютные измерения \( \beta \) излучающих радионуклидов

Дается описание детекторов \( 4 \pi \) Г. М. и приводятся некоторые их характеристики.

Отсутствие ложных импульсов показано путем измерения возрастания интервалов между импульсами.

Рассматриваются некоторые проблемы, относящиеся к поглощению фольги и источника при помощи \( \text{Sr}^{90} \) и \( \text{Y}^{90} \).

Представлен также экспериментальный метод с использованием парных источников, направленный на поправку исчисляемых потерь в фольге и до некоторой степени в источнике поглощения. Дается описание подготовки стандартов \( \text{Bi}^{210} \) (RaE).

Представлено примечание о проблеме потерь фольги от поглощения при сцинтилляционном абсолютном счете.
Sobre la medición absoluta de los emisores β

Se describen los detectores Geiger-Müller 4π y algunas de sus características.

La ausencia de impulsos parásitos se comprueba midiendo el ritmo a que se suceden los intervalos entre los impulsos.

Con ayuda de Sr90 y Y90, se estudian algunos de los problemas de autabsorción y de la absorción por las hojas.

Con el propósito de corregir la disminución del número de impulsos debido a la absorción en las hojas y, hasta cierto punto, a la autoabsorción, el autor propone un método experimental que emplea fuentes pareadas.

Asimismo, describe la preparación de patrones de Ba210 (RaE). La memoria termina con una nota sobre el problema de las pérdidas por absorción en las hojas en el recuento absoluto por centelleo.

1. Introduction

Although, in many applications of radionuclides, relative measurements are usually sufficient for the ends in view, it is also true that a precise knowledge of the disintegration rate of a radionuclide in a sample is of fundamental importance in nuclear physics and nuclear chemistry and some of their applications, and, in radiobiology, in special radiation therapy with radionuclides.

The purpose of this paper is to describe techniques and methods we have been using for the disintegration-rate determination, by 4π counting, of β-emitting radionuclides. Special reference will be made to 4π counting in the Geiger-Müller region.

The preparation and calibration of β standards of Bi210 (RaE) and α standards of Po210 (RaF) are described, including a chromatographic method for the separation of RaD, RaE and RaF and the measurement of the disintegration rate of the solutions by 4π counting in the GM region and/or 4π counting in the proportional region.

A scintillation method for the absolute α counting of Po210 is presented.

Finally reference is made to an absolute method of coincidence counting of radionuclides using sodium iodide (Thallium activated) crystal for the detection of the γ radiation and a plastic scintillation detector with the source between, which is evaporated on to very thin scintillation foils.

2. 4π counting detectors in the Geiger-Müller region

The 4π GM detectors in use in our laboratory follow, with some modifications, a model by N. Veall to which reference has been made in previous publications (20) (9).

Figure 1, shows one of the detectors. It consists essentially of a cylinder 4 cm in diameter divided across a diameter by the support of the source-mount. The latter is in brass as is the whole of the body of the detector and it consists of two pieces having a circular aperture 1.8 cm in diameter with the source-mount sandwiched between them.

The anodes are two 0.1 mm diameter wolfram wires which are stretched tight by the action of a terminal glass bead on a wolfram coil within the upper insulating glass tube.

202
The wolfram wires guided through the glass tubes are welded to the lower glass tube which is sealed to the brass cylinder with a radite. All the seals are covered with a lacquer layer in order to make the detector vacuum tight.

Constantan wires welded to the wolfram wires provide the electrical connections to the external circuits.

The gaseous filling of the detector is carried out through the upper part where a ground brass joint allows the connection or disconnection of the detector to the filling system.

Figure 1 shows a glass $4\pi$ detector whose features are very similar to the metallic one just described.

The gaseous filling of the detector is carried out through a standard pyrex glass ground joint.

The cathode is formed by coating the internal surface of the pyrex tube with colloidal graphite (Aquadag-Acheson Colloids). Polystyrene disks are the supports for the glass shielding tubes and the brass support of the source-mount. These disks are sealed to the glass with a concentrated solution of polystyrene in xylene.

Continuity of the source-mount support with the cathodic surface is assured by painting the polystyrene surfaces with colloidal graphite in alcohol (Acheson Colloids-Dag dispersion n° 80).

3. Counting efficiency of the $4\pi$ detector operating in the Geiger-Müller region

The occasional failure of a $\beta$ particle from a radionuclide source inside the $4\pi$ detector to produce an observable pulse arises mainly from the following causes:

a) The particle does not enter the sensitive volume of the counter because it is absorbed in the source-mount or absorbed in the material of the source itself or because the sensitive volume does not include the region where the source is deposited.
b) No production of an ion-pair in the counting gas. In all the types of 4π detectors used in our work a particle leaving the source has the possibility of travelling at least 2 cm inside the counter before arriving at the walls except for a very small region between the source and the circular aperture of the source-mount support. Even if 5% of the β radiation is emitted within this region the probability, for a normal gaseous filling, that a β particle does not produce an ion-pair in a 4 mm path through the gas in about 1%. The counting loss would then be about 0.1%.

In a previous paper (20) it was experimentally shown, by studying the way the counting rate of a P32 source varied with the diameter of the aperture of the source-mount support, that under the actual working conditions this loss was in fact negligible.

c) Possibility of the ion-pair not succeeding in triggering the counter. The probability is certainly low unless the electron attaches itself to a gaseous molecule, forming a negative ion, subsequently lost by recombination. Purity of the counting gas will eliminate this loss. The possibility that the discharge in the counter is not registered can be eliminated by a suitable choice of auxiliary electronic equipment.
d) Losses due to the resolving time of the counting assembly.

As a rule the resolving time is dependent, not on the detector but on the counting equipment and the losses are corrected by the well known expression

\[ N = \frac{N_0}{1 - N_0 \tau} \]

where \( N \) is the true counting rate, \( N_0 \) the observed counting rate and \( \tau \) the resolving time of the counting equipment. This resolving time was accurately checked. One simple method for this was the use of a pulse generator giving two equal pulses whose separation could be continuously varied. These pulses were injected at the input of the counting equipment and when the separation between the pulses became just less than the resolving time, they would be counted as one pulse only.

Another method for the same determination was to use one pulse whose frequency was continuously variable. If the pulse was injected into the counting equipment when the inverse of the frequency was greater than the resolving time, the pulse counting rate would double. We believe that the resolving time can be estimated to an accuracy of about 2%. So, for a counting rate of 10 000 counts per minute and a resolving time of 300 \( \mu \text{s} \), corresponding to a counting loss of 5%, this error of 2% in the resolving time corresponds to an inaccuracy of about 0.1% in the corrected counting rate.

Figure 3
4. Some counting characteristics of the 4π detectors

The length of the plateau is about 100 V and the latter can be considered statistically flat for the detectors used during this work. Sometimes, for an unknown reason, some of the detectors we build show plateaux with slopes of 1—2% per 100 volt and these are not used for very precise work.

After filling (usually 1 cm Hg of alcohol + 9 cm Hg of argon) we have to wait about 5 minutes for the stabilization of the operating conditions.

The filling system is shown in figure 3 where a 4π detector can be seen in position.

The Argon commercially available (Sociedade Portuguesa do Ar Liquido) is 99.9% pure (Oxygen — less than 0.005%, Nitrogen — less than 0.05%, Hydrogen — less than 0.001% and water — less than 10 mg/m²).

In some of the measurements the detector was connected to a “probe unit” (1014 A — Harwell) which, by lowering the anode potential by 240 V following a discharge, for the required resolving time, is intended to eliminate the spurious counts that could be caused by interaction of the positive cloud with the cathode during this time interval.

Nevertheless spurious counts can also be present: from the electronic connections; from negative ions capable of giving origin to pulses beyond the resolving time and from the positive cloud corresponding to the discharge of the negative ions.

Spurious counts could, of course, lead to an apparent efficiency greater than 100%.

The presence of flat plateaux points to the absence of spurious counts but we tried a more direct approach following a method used by PUTMAN (3) based on the interval distribution of random processes.

In radioactive decay the necessary conditions are fulfilled for the Poisson's distribution

\[ P_u = \frac{x^u}{u!} e^{-x} \]  

where \( p_u \) is the probability of observing \( u \) disintegrations when the average for a large number of observations is \( x \) disintegrations.

The probability that there will be no disintegrations in a time interval \( t \) if \( n \) is the average disintegration rate is

\[ p_0 = e^{-nt} \]  

The probability of a disintegration in the time interval \( \delta t \) is, of course, independent of \( t \) and is \( n\delta t \).

The combined probability of no disintegration in the time interval \( t \), but one between time \( t \) and time \( t + \delta t \) is, of course,

\[ dp = n e^{-nt} \delta t \]  

If we can measure the number of time intervals between disintegrations per unit time, varying \( t \) for a \( \delta t \) constant which is small compared with \( t \), we will have, calling this magnitude \( r(t) \)

\[ r(t) = n dp = n^2 e^{-nt} \delta t \]  

By using this formula (4.4) it is possible to detect the presence of spurious counts in the discharges of a detector.
The electronic circuit used for that determination is similar to the one described by Putman (3). Figure 4 is a block diagram of the electronic apparatus.

![Figure 4](image)

Figure 5 represents the results obtained with a Co$^{60}$ source within a $4\pi$ Geiger-Müller detector giving 12,730 counts per minute, connected to a "probe unit" with a resolving time of 300 $\mu$s.

![Figure 5](image)

From equation (4.4) we see at once that

$$\frac{d \ln r}{dt} = -n$$

(5.4)

The value of $n$ calculated from expression (5.4) is 13,400 counts per minute which is 5.4% higher than the figure experimentally obtained.

In a previous paper (22) reference was made to the fact that with $^{32}$P sources, when the minimum path for the $\beta$ particles was reduced to about 2.5 cm, a decrease of 4% in the counting rate was observed with a normal filling (1 cm alcohol + 9 cm argon). The difference disappeared if the pressure was increased to 20 cm.

This led us to investigate the influence of pressure on the counting rate. Figure 6 shows the high voltage characteristics of the $4\pi$ detector. When the alcohol pressure was maintained at 1 cm Hg, the argon pressure went...
up from 2 cm Hg to 25 cm Hg. It will be seen that the plateau is flat and the counting rate variation was statistically quite acceptable.

5. Experimental procedures and results

A. Generally the problem is to measure the specific disintegration rate of a given solution and this leads us very often to make successive dilutions of the primary solution until we get a final solution with a specific radioactivity of 0.2—0.3 μc/g. Every precaution must be taken to guard against errors introduced in each dilution and against losses of the radioactive material by adsorption in the walls of the containers.

To decrease the importance of this effect one of the precautions is to treat the walls of the container with a silicone solution and to cure the resulting coating at about 300° C for a few hours. HOUTERMANS et al. (8) suggested for the same purpose a coating with dimethyldichlorosilane.

Even with all these precautions, however, it is practically impossible to avoid losses with carrier-free radioactive solutions and it is always necessary
to add to the solution to be calibrated, the stable isotope of the radionuclide being studied. The quantity added however must be kept to a minimum (generally about 50\(\mu g/cm^3\)) in order not to increase the self-absorption effect but this correction is not necessarily proportional to the solid content of the source. It has been shown \(^\text{(12)}\) that we cannot reduce the self-absorption losses below a certain amount, but we can at least decrease the number of "islands" formed consisting in aggregates of crystals, without changing essentially their size which is the important factor depending greatly on the methods of source preparation \(^\text{(24)}\).

Another very important precaution is to maintain the pH in all the solutions at a value which guarantees their chemical stability.

From the final solution with the desired radioactivity concentration, the sources are prepared by evaporating under infra-red irradiation, a certain amount of the solution, determined by weighing the pyknometer shown in figure 7 before and after delivering the solution drops.

B. Source-mount corrections

\(^{(i)}\) As pointed out above, since the source-mount has a finite thickness and a continuous distribution of \(\beta\) energies from zero to a certain end-point energy is present in the source to be measured, there is always absorption in the source-mount, increasing with decreasing \(\beta\) end-point energy, as is well known.

Since in this as in all similar situations it is better to eliminate the loss than to assess its value, considerable effort has been made by several investigators to work out methods for the production of very thin source-mounts.

Recently a method has been reported for the successful fabrication of films with superficial densities below 1 \(\mu g/cm^2\) using a polyvinyl-chloride-acetate copolymer (VYNS resin) with satisfactory mechanical and chemical characteristics \(^\text{(16)}\).

Since the plastic film used must be essentially at cathode potential it must be made conducting by a suitable coating, usually a metallic one, applied by sputtering or distillation in vacuo. If the film has a superficial density of 5—10 \(\mu g/cm^2\) the losses in the source-mount for most ordinary work can be considered negligible.

However, preparation and control of the films is a lengthy procedure and although recognising our limitations we have been using aluminium foils which can be obtained in superficial densities as low as 160 \(\mu g/cm^2\) (Geo M. Whiley, Ruislip, Middx, England). The mechanical stability is quite good but the aluminium does not show very good chemical resistance to acids or alkalis which imposes limitations on the nature of the solutions to be calibrated.

Methods of determining the source-mount correction have been described in the literature: the sandwich method \(^\text{(7)}\), calculation from 2\(\pi\) and 4\(\pi\) counting rates \(^\text{(10)}\) \(^\text{(6)}\) \(^\text{(18)}\) and the use of absorption curves \(^\text{(11)}\) \(^\text{(18)}\). A review and criticism of all these methods has been recently published \(^\text{(18)}\).

In a previous publication we have made a study of the variation of the counting rate in accordance with the thickness of the source-mount. The curves shown in figure 8 and figure 9 represent some of the results we obtained with \(\text{I}^{131}\), \(\text{Na}^{22}\), \(\text{Co}^{60}\) and \(\text{Sr}^{90} + \text{Y}^{90}\).
Extrapolation of the curves to zero thickness will give the wanted correction. However, as was suspected previously (11) this extrapolation could not be accurate below the smallest thickness of the aluminium foils available and this was demonstrated by other authors (18) who believe that, provided very thin films are available, this procedure will give a more accurate value for the source-mount correction.

It is perhaps interesting to notice the trend of the curve corresponding to Na$^{22}$ as compared to the one obtained for I$^{131}$ in figure 8. An identical source-mount correction for both radionuclides would be expected since some data (14) show that the absorption of Na$^{22}$ positrons is the same as that of I$^{131}$ electrons up to 90 mg/cm$^2$ of aluminium absorbers. However, figure 8 points to a negligible correction for the positrons of Na$^{22}$ in aluminium foils 0.2 mg/cm$^2$ thick, although a correction of 2% for the electrons of I$^{131}$ is indicated.

This result was confirmed by another procedure as shown below and perhaps could be explained by Coulomb forces at play.
(ii) The two paired-sources method for the source-mount absorption correction. In order to correct for the non-uniformity of the aluminium foils since the superficial density measured is an average for the whole foil and also eliminate the need for determinations of each foil thickness, we suggested a very simple method for the calculation of the losses due to absorption in the source-mount.

Let $K$ be the specific radioactivity (measured in counts per minute per milligram) of a radioactive solution to be calibrated. If from this solution we prepare, as described above, two sources, source I with a mass of $m_1$ milligrams and source II with a mass of $m_2$ milligrams, we should have, if there were no losses in the $4\pi$ counter

Source I: $N_1^0 = m_1 K$

Source II: $N_2^0 = m_2 K$

If we call $\alpha = \frac{m_2}{m_1}$

then $N_2^0 = \alpha N_1^0$.

With the sources within the detector we obtain

$$N_1 = \frac{N_1^0}{2} + \frac{N_1^0}{2} (R_1 + T_1)$$

where

$R_1 =$ the source-mount back-scattering coefficient

$T_1 =$ the transmission of the film to the incident $\beta$ radiation from source I.

Identically, for the source II

$$N_2 = \alpha \frac{N_1^0}{2} + \alpha \frac{N_1^0}{2} (R_2 + T_2)$$

Counting the two sources together, placed face to face, we shall have

$$N_{1,2} = T_1 \frac{N_1^0}{2} (1 + R_2) + T_2 \frac{N_1^0}{2} (1 + R_1) +$$

$$+ \alpha T_1 \frac{N_1^0}{2} (1 + R_2) + T_2 \frac{N_1^0}{2} (1 + R_1)$$ assuming the same transmission coefficient for the incident $\beta$ radiation and for the back-scattered $\beta$ radiation.

From these equations we have

$$N_1^0 = \frac{N_1}{1 + R_1} + \frac{N_2}{\alpha (1 + R_2) - \frac{N_{1,2}}{(1 + \alpha) (1 + R_1) (1 + R_2)}}$$

Neglecting second-order terms we can write,

$$N_1^0 = N_1 + \frac{N_2}{\alpha} - \frac{N_{1,2}}{1 + \alpha}$$

which will be used in a certain number of experiments.
Tables I, II and III show results obtained applying this method to $\text{Co}^{60}$, $\text{S}^{35}$ and $\text{Sr}^{90} + \text{Y}^{90}$ sources on source-mount of various superficial density values.

**TABLE I**
The two paired-sources method applied to $\text{Co}^{60}$ sources

<table>
<thead>
<tr>
<th>Sup. density mg/cm$^2$</th>
<th>Source mass mg</th>
<th>N cps</th>
<th>$N_{12}$ cps</th>
<th>K cps/mg</th>
<th>Correct. %</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.198</td>
<td>41.77</td>
<td>257.7</td>
<td>450.8</td>
<td>6.45</td>
<td>4.5</td>
</tr>
<tr>
<td>0.198</td>
<td>35.83</td>
<td>218.4</td>
<td></td>
<td>6.45</td>
<td>5.9</td>
</tr>
<tr>
<td>0.205</td>
<td>38.99</td>
<td>240.2</td>
<td>447.2</td>
<td>6.49</td>
<td>5.3</td>
</tr>
<tr>
<td>0.204</td>
<td>38.42</td>
<td>234.6</td>
<td></td>
<td>6.49</td>
<td>6.3</td>
</tr>
<tr>
<td>0.184</td>
<td>24.94</td>
<td>149.7</td>
<td>391.7</td>
<td>6.44</td>
<td>7.3</td>
</tr>
<tr>
<td>0.698</td>
<td>46.85</td>
<td>276.3</td>
<td></td>
<td>6.44</td>
<td>9.2</td>
</tr>
</tbody>
</table>

The cobalt sources were prepared by precipitation of $\text{CoCl}_2$ solution with 0.5 N Ammonia.

**TABLE II**
The two paired-sources method applied to $\text{S}^{35}$ sources

<table>
<thead>
<tr>
<th>Sup. density mg/cm$^2$</th>
<th>Source mass mg</th>
<th>N cps</th>
<th>$N_{12}$ cps</th>
<th>K cps/mg</th>
<th>Correct. %</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.221</td>
<td>28.28</td>
<td>181.7</td>
<td>357.8</td>
<td>7.09</td>
<td>10.4</td>
</tr>
<tr>
<td>0.190</td>
<td>32.17</td>
<td>211.8</td>
<td></td>
<td>7.09</td>
<td>7.7</td>
</tr>
<tr>
<td>0.186</td>
<td>31.16</td>
<td>201.1</td>
<td>406.0</td>
<td>6.86</td>
<td>6.4</td>
</tr>
<tr>
<td>0.202</td>
<td>37.40</td>
<td>236.7</td>
<td></td>
<td>6.86</td>
<td>8.4</td>
</tr>
<tr>
<td>0.643</td>
<td>36.86</td>
<td>215.4</td>
<td>305.0</td>
<td>7.02</td>
<td>20.1</td>
</tr>
<tr>
<td>0.539</td>
<td>26.17</td>
<td>157.4</td>
<td></td>
<td>7.02</td>
<td>16.8</td>
</tr>
<tr>
<td>0.614</td>
<td>40.89</td>
<td>234.2</td>
<td>338.3</td>
<td>6.80</td>
<td>18.7</td>
</tr>
<tr>
<td>0.485</td>
<td>28.40</td>
<td>169.4</td>
<td></td>
<td>6.80</td>
<td>14.4</td>
</tr>
</tbody>
</table>

Carrier-free $\text{S}^{35}$ with traces of $\text{H}_2\text{SO}_4$, pH = 2; 0.1 $\mu$g sol./mg.

**TABLE III**
The two paired-sources method applied to $\text{Sr}^{90} + \text{Y}^{90}$ sources

<table>
<thead>
<tr>
<th>Sup. density mg/cm$^2$</th>
<th>Source mass mg</th>
<th>N cps</th>
<th>$N_{12}$ cps</th>
<th>K cps/mg</th>
<th>Correct. %</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.168</td>
<td>32.41</td>
<td>257.5</td>
<td>486.1</td>
<td>8.06</td>
<td>1.6</td>
</tr>
<tr>
<td>0.168</td>
<td>31.50</td>
<td>243.3</td>
<td></td>
<td>8.06</td>
<td>4.4</td>
</tr>
<tr>
<td>0.196</td>
<td>32.46</td>
<td>278.6</td>
<td>457.6</td>
<td>8.01</td>
<td>0.8</td>
</tr>
<tr>
<td>0.726</td>
<td>37.39</td>
<td>288.0</td>
<td></td>
<td>8.01</td>
<td>4.2</td>
</tr>
<tr>
<td>0.191</td>
<td>38.43</td>
<td>224.3</td>
<td>482.3</td>
<td>8.06</td>
<td>2.1</td>
</tr>
<tr>
<td>0.201</td>
<td>34.57</td>
<td>270.3</td>
<td></td>
<td>8.06</td>
<td>3.2</td>
</tr>
</tbody>
</table>

With the $\text{Na}^{22}$ we prepared three sources and measured the corresponding counting rates. Next we grouped the sources two by two and measured the two sources together. The results obtained are shown in Table IV.
### TABLE IV

<table>
<thead>
<tr>
<th>Source</th>
<th>Sup. density (mg/cm²)</th>
<th>N (cps)</th>
<th>Nₐ, b (paired counting rate) (cps)</th>
<th>Nₐ + Nₐ (Adding the ind. counting rates)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.206</td>
<td>143.3</td>
<td>(1,2) 285.4</td>
<td>285.4</td>
</tr>
<tr>
<td>2</td>
<td>0.182</td>
<td>142.1</td>
<td>(1,3) 276.2</td>
<td>276.1</td>
</tr>
<tr>
<td>3</td>
<td>0.186</td>
<td>132.8</td>
<td>(2,3) 274.0</td>
<td>274.9</td>
</tr>
</tbody>
</table>

These results which are in agreement with the data plotted in figure 8, lead to a negligible correction for the positrons of Na²² in 0.2 mg/cm² aluminium foils. For I¹³¹ we get a foil correction of about 3.0% for the aluminium foils with the same superficial density.

Although the two paired-sources method, because of the simplifications made during the derivation of the final expression (6, 5.2.2.) — namely the assumption of the same transmission for the β particles incident on the source-mount and for the scattered radiation —, cannot be completely justified on theoretical grounds and is even, accepting some published data (19), vulnerable to the criticism made on the "sandwich" method, it is nevertheless true that results presented above for low energy β emitters such as Co⁶⁰ (312 keV) and S³⁵ (167 keV) show a noticeable internal consistency even for source-mounts of a superficial density of about 0.6 mg/cm².

(iii) Self-absorption and source-mount losses for Sr⁹⁰ sources. The fact that the daughter of Sr⁹⁰ (end-point energy — 545 keV) is Y⁹⁰ (end-point energy — 2260 keV), a radionuclide for which we can possibly assume that the self-absorption losses and the source-mount losses are negligible, leads to a simple way of studying these corrections for the Sr⁹⁰.

Suppose that we completely separate the strontium from the yttrium and prepare a source to be measured in a 4π detector.

If the measurement is made a certain time t after the separation, then, when

\[ \frac{\tau}{N_s} = \text{the fraction of the disintegration rate of the Sr⁹⁰ source} \]
\[ N_s = \text{counts in the 4π detector due to self-absorption and source-mount losses} \]
\[ \alpha(t) = N_y(t)/N_s \text{ where } N_y \text{ is the disintegration rate of the Y⁹⁰ existent at time } t \text{ after separation} \]
\[ T_s = 1.011 \times 10^4 \text{ days (the half-life of Sr⁹⁰)} \]
\[ T_y = 2.675 \text{ days (the half-life of Y⁹⁰)} \]
\[ \lambda_y = 0.2591 \text{ days}^{-1} \text{ (disintegration constant of Y⁹⁰)} \]

the following expression is obtained for the counting rate at time t₁ after the separation,

\[ N_1 = \tau N_s + \alpha(\tau_1) N_s \]

and at time t₂, identically,

\[ N_2 = \tau N_s + \alpha(\tau_2) N_s \]

where

213
\[ z(t) = \frac{T_s}{T_s - T_y} \left(1 - \exp \left(-\frac{T_s}{T_s - T_y} \lambda_y t\right)\right) \]

or, substituting values,
\[ \alpha(t) = 1000 [1 - \exp(-0.2591 t)]. \]

From the expressions above,
\[ \tau = \frac{\alpha(t_1)N_2 - \alpha(t_2)N_1}{N_1 - N_2} \]

and the total losses will be
\[ \sigma = 1 - \tau. \]

The aluminium foils used had a superficial density close to 0.2 mg/cm². Table V shows the data obtained.

**TABLE V**

<table>
<thead>
<tr>
<th>Source No</th>
<th>t days</th>
<th>Source mass mg</th>
<th>Specific act. cps/mg</th>
<th>( \alpha(t) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.104</td>
<td>24.25</td>
<td>3.56</td>
<td>0.0265</td>
</tr>
<tr>
<td>2</td>
<td>0.144</td>
<td>25.24</td>
<td>3.57</td>
<td>0.0365</td>
</tr>
<tr>
<td>3</td>
<td>0.165</td>
<td>25.75</td>
<td>3.55</td>
<td>0.0418</td>
</tr>
<tr>
<td>4</td>
<td>0.183</td>
<td>21.24</td>
<td>3.60</td>
<td>0.0464</td>
</tr>
<tr>
<td>5</td>
<td>0.243</td>
<td>30.16</td>
<td>3.64</td>
<td>0.0661</td>
</tr>
<tr>
<td>6</td>
<td>0.271</td>
<td>27.35</td>
<td>3.65</td>
<td>0.0678</td>
</tr>
<tr>
<td>1</td>
<td>19.06</td>
<td>13.32</td>
<td>6.92</td>
<td>0.994</td>
</tr>
<tr>
<td>2</td>
<td>19.08</td>
<td>19.53</td>
<td>7.00</td>
<td>0.994</td>
</tr>
<tr>
<td>3</td>
<td>19.09</td>
<td>22.03</td>
<td>6.95</td>
<td>0.994</td>
</tr>
<tr>
<td>4</td>
<td>19.10</td>
<td>14.28</td>
<td>6.98</td>
<td>0.994</td>
</tr>
<tr>
<td>5</td>
<td>19.12</td>
<td>17.75</td>
<td>6.95</td>
<td>0.994</td>
</tr>
<tr>
<td>6</td>
<td>19.14</td>
<td>14.04</td>
<td>6.92</td>
<td>0.994</td>
</tr>
</tbody>
</table>

Grouping the sources with the same number we obtain

<table>
<thead>
<tr>
<th>Source No</th>
<th>Total efficiency ( \tau )</th>
<th>Total losses ( \sigma )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.997</td>
<td>0.003</td>
</tr>
<tr>
<td>2</td>
<td>0.959</td>
<td>0.041</td>
</tr>
<tr>
<td>3</td>
<td>0.950</td>
<td>0.050</td>
</tr>
<tr>
<td>4</td>
<td>0.961</td>
<td>0.039</td>
</tr>
<tr>
<td>5</td>
<td>0.961</td>
<td>0.039</td>
</tr>
<tr>
<td>6</td>
<td>0.966</td>
<td>0.033</td>
</tr>
</tbody>
</table>

Average — 0.966  Average — 0.033

This will give, for the 4 \( \pi \) measurements of \( \text{Sr}^{90} + \text{Y}^{90} \) sources, a correction of 1.7\%.
6. Secondary system for the absolute measurement of $\beta$-emitting radionuclides

It is useful to have a system of known efficiency for the different radionuclides in frequent use which is of simple operation and good stability. We have been using for the past four years (22) a set-up which satisfies those requirements.

The counting assembly consisting essentially of two end-window Geiger-Müller detectors in a rigid frame is shown in figure 10.

![Figure 10](image)

![Figure 11](image)
Separation between the two counters was chosen on the base of very simple calculations whose results are given in figure 11 which shows how the counting rate varies with the distance of a point source from the window of one of the detectors. The corresponding values for one detector only and the counter aperture for different separations of the detectors and a point source midway between them are also given.

The source-mount has the same dimensions as the one used in the 4 $\pi$ detectors so that the same source measured in the 4 $\pi$ counter can be used for the calibration of the system.

The constancy of the response of the set-up for lateral displacement from the axis of the detectors up to 0.7 cm was experimentally checked.

From the efficiency values determined for several radionuclides a calibration curve was plotted (figure 12), showing the efficiency in terms of the end-point energy of the $\beta$ radiation of the radionuclides. From the values obtained for Co$^{60}$, P$^{32}$, and Au$^{198}$ where a single $\beta$ transition is observed and from the observed efficiency for the complex $\beta$ spectrum of I$^{131}$ and the Sr$^{80}$ + Y$^{90}$ sources, it was possible to assign more values to the curve as the cut-off energy for the superficial densities of the GM windows is 0.050 MeV.

We believe that for radionuclides with $\beta$ end-point energies from 0.3 to 2.3 MeV we can get an accuracy of about 5% by measurements with the device just described.

7. Preparation and calibration of $\beta$ standards of Bi$^{210}$ (Ra E) and $\alpha$ standards of Po$^{210}$ (Ra F)

A. For many applications of radioactive substances it is quite convenient to have solutions of long-lived radionuclides whose activity is well known and of desirable energies. For that reason some work has been done on the preparation and measurement of $\beta$ standards of Bi$^{210}$ and $\alpha$ standards of Po$^{210}$ both present in old radon seeds.

In effect, equilibrium is established between the radionuclides Ra D (Pb$^{210}$), Ra E (Bi$^{210}$) and Ra F (Po$^{210}$) after 30 days and, as the Po$^{210}$ has a half-life of 19.4 years, we can use a solution containing Ra D as standard $\beta$ of Ra E taking the precaution after equilibrium is established, of covering the source with a foil of superficial density greater than 6 mg/cm$^2$, enough
to stop the $\beta$ radiation of Pb$^{210}$ [end-point energy (15%)—61 keV] and the $\alpha$ radiation of Po$^{210}$ (5.303 MeV).

If we determine the efficiency of a $\beta$ detection device, like the one described above, with an absolute-measured Bi$^{210}$ source covered with a suitable foil, we can calibrate a given Ra D, E, F solution.

The preparation and calibration of Bi$^{210}$ sources has been reviewed by NOVEY (5). The following methods have been used:

1— Calculation of the $\beta$ disintegration rate of Ra E from the rate of growth of the $\alpha$ activity of a pure Ra D source (2).

2— Calculation of the $\beta$ activity from the rate of growth of $\alpha$ activity of a pure Ra E source.

3— Calculation of the Ra E activity from a knowledge of the radium content of a pitchblende sample from which Ra D and stable lead are quantitatively removed electrolytically (4).

4— Calculation of the Ra E activity for sources prepared from a solution of Ra D, E, F in equilibrium by measuring the $\alpha$ disintegration rate (6).

The method chosen for this work was to prepare pure or almost pure solutions of Ra E and count the $\beta$ plus $\alpha$ activity in a 4 $\pi$ detector. From this data the $\beta$ disintegration rate can be calculated (20).

B. In a first series of experiments the separation of Ra E from Ra D and Ra F was done using ordinary chemical and electrochemical procedures.

To the solution obtained by treating old radon seeds, broken to pieces, with hydrochloric acid 0.1 N, about 20 mg of nickel in small metallic foils was added. Agitation during heating to 80°C for 3 minutes was performed. In this way the electrochemical displacement of Ra F and Ra E occurs. The nickel separated by centrifugation was taken with H Cl, 0.1 N and the whole procedure repeated again and again.

In the next step the nickel was dissolved in aqua-regia and a solution in distilled water prepared (about 10 cm$^3$).

Bismuth nitrate was added at this stage and bismuth hydroxide was precipitated with ammonia. The pp. contains Ra E and Ra F.

Dissolution of the bismuth hydroxide with H Cl, 0.1 N was next carried out and the Ra F displaced from the solution with silver foils in constant agitation within the solution for 24 hours.

As is well known, the complete separation of Ra E from Ra F is very difficult to obtain by chemical and electrochemical methods so it is to be expected that the final solution contains both radionuclides. For that reason we try to detect simultaneously the $\beta$ particles of Bi$^{210}$ and the $\alpha$ particles of Po$^{210}$ in our 4 $\pi$ detector.

From the end-point energy of the electrons of Bi$^{210}$ (1.155 MeV) and the $\alpha$ energy (5.303 MeV) we would expect an identical source-mount correction of about 29% (for aluminium foils of superficial density of 0.2 mg/cm$^2$).

If $N(t)$ is the disintegration rate measured at time t after the separation of Ra E from Ra F, due to a mixed source of these radionuclides,
B(o) — is the disintegration rate corresponding to the β radiation of Ra E, and

A(o) — is the disintegration rate corresponding to the α radiation of Ra F present in the source at time t = 0,

then

\[ N(t) = B(o) \exp(-\lambda_E t) \left( \frac{2 - \exp(\lambda_F - \lambda_E)t}{1 - \lambda_E/\lambda_F} \right) + A(o) \exp(-\lambda_F t) \]

Measuring N(t) at two instants t₁ and t₂, we can calculate by this expression B(o) and A(o).

Since we started with a chlorhidric solution we have to neutralize the solution with ammonia in order not to destroy the aluminium source-mounts.

It was at once clear that we no longer had a flat plateau and so the counting rate for any particular source had to be taken as the value corresponding to the threshold of the plateau derived graphically by carefully extrapolating the linear curve obtained. This particular point could be accurately determined, if the initial part of the high voltage characteristic, just before the plateau, was carefully plotted.

Further study of the problem leads us to the conclusion that this slope of the plateau was due, at least in part, to the solid content of the source.

Working with carrier-free sources, this problem almost disappears.

Table VI shows that the results are quite satisfactory. The last column contains the calculated values using an average value for the β specific counting rate at time t = 0, B(o).

**TABLE VI**

**Determination of the β activity of Bi²¹⁰ (RaE)**

<table>
<thead>
<tr>
<th>Measurement time days</th>
<th>Specific counting rate cpm/mg</th>
<th>Spec. counting calc. for B(o) = 105.8 cpm/mg</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.151</td>
<td>122.0</td>
<td>122.3</td>
</tr>
<tr>
<td>0.167</td>
<td>124.0</td>
<td>112.8</td>
</tr>
<tr>
<td>1.043</td>
<td>113.4</td>
<td>89.9</td>
</tr>
<tr>
<td>1.063</td>
<td>110.9</td>
<td>81.9</td>
</tr>
<tr>
<td>3.052</td>
<td>92.4</td>
<td>56.5</td>
</tr>
<tr>
<td>3.125</td>
<td>81.4</td>
<td>56.5</td>
</tr>
<tr>
<td>4.091</td>
<td>81.4</td>
<td></td>
</tr>
<tr>
<td>4.920</td>
<td>56.5</td>
<td></td>
</tr>
<tr>
<td>8.069</td>
<td>56.5</td>
<td></td>
</tr>
<tr>
<td>8.080</td>
<td>56.5</td>
<td></td>
</tr>
</tbody>
</table>

From the data obtained it was calculated that at t = 0 the α radioactivity from Ra F present in the solution corresponds to 16.1% of the total disintegration rate.

C. A more simple approach to this problem was to make successive determinations measuring the α activity in the proportional region and next the β plus α activity in the Geiger-Müller region. Our 4π detectors with an
argon filling at atmospheric pressure show very flat plateaux for a counting in the proportional zone and so, with due regard for the solid content of the source, the final result can be obtained with all the accuracy desired.

D. Separation of RaD, E, F by paper chromatography. Calibration of pure RaE (Bi$^{210}$) solutions.

With the usual chemical and radiochemical methods it is extremely difficult to obtain pure RaE solutions. Chromatography, as shown by the results recently published, is a much better and simpler method for this purpose.

LEDERER (21) used ascending paper chromatography for the separation of RaD, E, F in 0.1 N HCl solution with Whatman paper No 1 and the development phase was butanol saturated with HCl, 0.1 N.

We have found (26) that some improvement in the separation of RaF from RaE can be obtained using descending paper chromatography with Whatman paper No 1 and tertiary amyl alcohol saturated with HCl, 0.1 N as the development phase. Table VII shows the values of R observed with the two systems.

TABLE VII
Separation of RaD, E, F by paper chromatography

<table>
<thead>
<tr>
<th>System</th>
<th>Development phase</th>
<th>RaD $R_f$</th>
<th>RaE $R_f$</th>
<th>RaF $R_f$</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Whatman 1, ascending</td>
<td>Butanol saturated HCl, 0.1 N</td>
<td>0.148</td>
<td>0.574</td>
<td>0.833</td>
<td>(21)</td>
</tr>
<tr>
<td>Whatman 1, descending</td>
<td>Tertiary amyl alcohol sat. HCl, 0.1 N</td>
<td>0.168</td>
<td>0.480</td>
<td>0.798</td>
<td>(26)</td>
</tr>
</tbody>
</table>

Figure 13 shows a radiochromatogram and the corresponding autoradio-graph obtained by the amyl alcohol technique. Developing time was 16 hours.

The good separation obtained ought to allow us to prepare pure carrier-free solutions of the three radionuclides.
The purity of solutions obtained directly from the paper chromatogram was checked for the Ra F (Po\(^{210}\)) by simply measuring sources in the proportional and the Geiger-Müller region as described above.

For the Ra E (Bi\(^{210}\)) the same method could be used. The problem can also be solved quite simply by working only on the Geiger-Müller region.

If we start with a pure Ra E source using the same nomenclature

\[ N(t) = B(t) + A(t) \]

where

\[ B(t) = B(0) \exp(-\lambda_E t) \]

Also, since Ra F is the daughter of Ra E,

\[ A(t) = B(0) \frac{\exp(-\lambda_E t) - \exp(-\lambda_F t)}{1 - \lambda_F / \lambda_E} \]

From this expression we can derive

\[ F(t) = \frac{B(0)}{N(t)} = \frac{(\lambda_E / \lambda_F) - 1}{(\lambda_E / \lambda_F - 2) \exp(-\lambda_E t) + \exp(-\lambda_F t)} \]

For

\[ \lambda_E = 1.377 \times 10^{-1} \text{ d} \]

\[ \lambda_F = 5.007 \times 10^{-3} \text{ d} \]

\[ F(t) = \frac{26.6}{25.6 \exp(-\lambda_E t) + \exp(-\lambda_F t)} \]

This function is represented in figure 14.

![Figure 14](image)

Table VIII shows a test of purity of a Ra E sample where we can see that the formula

\[ N(t) \times F(t) = B(0) \]

remains fairly constant.
TABLE VIII

Purity test of a RaE (Bi$^{210}$) source obtained from a paper chromatogram of RaD, E, RaF

<table>
<thead>
<tr>
<th>Time of meas. d</th>
<th>F(t)</th>
<th>B(α) = N(t) × F(t) c. p. m.</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.25</td>
<td>1.0168</td>
<td>6820</td>
</tr>
<tr>
<td>1.145</td>
<td>1.165</td>
<td>6848</td>
</tr>
<tr>
<td>2.063</td>
<td>1.310</td>
<td>6788</td>
</tr>
<tr>
<td>4.181</td>
<td>1.735</td>
<td>6750</td>
</tr>
<tr>
<td>6.000</td>
<td>2.184</td>
<td>6810</td>
</tr>
</tbody>
</table>

E. Absolute measurement of α emitters (Po$^{210}$) with 4π scintillation detectors.

The 4π gaseous detector working in the proportional region (17) or in the Geiger-Müller region (20) is a suitable apparatus for the absolute disintegration rate measurement of α emitters.

Work has been published on the absolute measurement of α emitters with 4π liquid scintillators (21) (13), where the radionuclide is incorporated in the liquid scintillator.

The technique we are going to describe is also very simple and uses the principle of the 4π solid scintillator counters.

The set-up is represented in figure 15.

![Figure 15](source.png)

The source is sandwiched between two thin scintillating foils. The filter paper covering the foils acts as reflector. The lead weight produces a good contact for the foils and with the help of a drop silicone oil ensures a good optical coupling with the window of the photomultiplier.

The scintillation foils are prepared following a method previously described. From a plastic scintillator obtained by polymerization of styrene containing 1% of p-therphenyl, 3% of POPOP, 3% of sodium stearate, for 24 hours at 130—140° C, we obtain, when the plastic is machined, chips of this material which we dissolve in toluene.

From a concentrated solution of this plastic material the foils are prepared in a very simple way by pouring a certain amount on a glass surface and, after obtaining an even layer, evaporating under infra-red irradiation. The perfectly transparent foils separate quite easily from the glass mounting.
For this particular purpose we use foils with a superficial density of 10 mg/cm$^2$.

Figure 16 shows the integral pulse counting rate obtained with a Po$^{210}$ source evaporated onto one of the plastic foils with an area of 4 cm$^2$, as a function of the discrimination voltage. As we see, the extrapolation correction for the counting rate obtained with 5 volts discrimination is only 1.2%.

The agreement between measurements made by this procedure and by using the 4$\pi$ GM detector is quite good, allowing for the small correction of about 2% for the $\alpha$ of Po$^{210}$ in aluminium foils of a superficial density of 0.2 mg/cm$^2$.

![Figure 16](image)

8. An experimental arrangement for the absolute measurement of radionuclides by the $\beta$-$\gamma$ coincidence method using solid scintillation detectors

The $\beta$-$\gamma$ coincidence technique has been shown by many workers to be a suitable procedure for the absolute measurement of a radionuclide where the $\beta$ emission is followed by a $\gamma$ transition \(^{(19)}\).

The method is limited by the various corrections which must be applied in order to get the final result.

The advantages arise from the fact that the absolute disintegration rate given by

$$N = \frac{N_{\beta} N_{\gamma}}{N_c}$$

where $N_{\beta}$, $N_{\gamma}$ and $N_c$ are the observed counting rates corrected for the background and for accidental coincidences in the $\beta$, $\gamma$ and coincidence channels, is independent of the counting efficiencies of the detectors and so independent of the source-mounts and self-absorption losses. As an independent way of assessing these losses its utilization is of importance in checking the results obtained with the 4$\pi$ methods.

Improvement of the technique can be expected mainly by way of improvement in the counting efficiency of the detectors and in the electronic characteristics of the associated equipment. We have concentrated on the first of these tasks.

Recently, very valuable work has been done on the theoretical and experimental aspects of the method \(^{(25)}\).
It can be seen that most of the corrections need only be quite small if consideration is given to the counting arrangement.

The more important corrections to be applied can be summarized as follows:

1. Corrections for the finite size of the source.

It was shown by Putman that the strict validity of the method can only be assumed if one of the detectors is equally sensitive to all parts of the source. If one of the detectors is almost 100% efficient no correction will be needed for this factor.

2. Correction for the $\gamma$ sensitivity of the $\beta$ detector, for $\gamma-\gamma$ coincidences, for the $\beta$ sensitivity of the $\gamma$ detector and for bremsstrahlung effects.

For the first factors CAMPION \textsuperscript{(25)} arrived at a correction given by

$$1 + \left( \frac{1 - \varepsilon_\beta}{\varepsilon_\beta} \right) \left[ (\varepsilon_\beta) \gamma - \varepsilon_\gamma / \varepsilon_\gamma \right]$$

where $\varepsilon_\beta$ is the efficiency of the $\beta$ detector, $\varepsilon_\gamma$ the efficiency of the $\gamma$ detector, $(\varepsilon_\beta) \gamma$ the sensitivity of the $\beta$ detector for the $\gamma$ radiation of the radionuclide and $\varepsilon_\gamma$ the probability that a coincidence will be observed when the $\beta$ particle is not counted.

Here again we see that a high efficiency for $\beta$ and $\gamma$ counting will make this correction quite small and that the same is true for the bremsstrahlung correction, since, for a $\beta$ counter with almost 100% efficiency, this radiation only increases the apparent sensitivity of the $\gamma$ detector. As for the $\beta$ sensitivity of the $\gamma$ detector it is quite simple, by suitable design, to prevent the $\beta$ particles from entering the $\gamma$ detector.

3. Corrections for the presence of $\beta-\gamma$ angular correlations.

This correction is in most cases negligible if the $\beta$ detector is 100% efficient.


This point can be covered by careful selection of the electronic equipment.

Taking into consideration the first three groups of corrections we suggested \textsuperscript{(25)} a coincidence counting arrangement which is shown in greater detail in figure 17.

![Figure 17](image-url)
The γ detector is a sodium iodide (Thallium activated) well-type crystal, the well being 4 cm deep and of 1.8 cm diameter.

The β detector is a double-cylinder shaped plastic scintillator divided across one diameter into two parts between which is sandwiched a source, evaporated-on and covered with a very thin plastic foil, prepared as described above (20).

The upper cylinder enters the well which has a radius (0.8 cm) greater than the maximum range of most of the β-emitting radionuclides to be measured.

Optical contact is made with silicone oil and the action of a metallic coil presses the two photomultipliers against the faces of the scintillators.

Figure 18 shows a photograph of the assembly with part of the lead shield removed.

![Figure 18](image_url)

**REFERENCES**

II. 2. 4. THE ABSOLUTE COUNTING OF NUCLIDES DECAYING BY $\beta$ EMISSION ONLY

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Abstract

In the determination of absolute disintegration rates of sources using the $4\pi \beta$ counter method it is difficult to measure or even estimate the self-absorption correction with any degree of certainty. The results support the view that the method of progressively diluting a stock solution until a constant activity is obtained only minimizes self-absorption and does not eliminate it. Further, any extrapolation technique based on a plot of apparent activity against mean source superficial density is not recommended because of the large scatter of the results.

As a first approach to the problem therefore we have measured, using the $4\pi \beta\gamma$ coincidence technique, the self-absorption as a function of $\beta$-ray end-point energy for a constant mean source superficial density of 2 $\mu$g/cm$^2$ and for a number of source-preparation techniques. The self-absorption varied from a few tenths of 1% at $\beta$ end-point energies of 1.39 MeV to about 20 to 40% at 0.086 MeV, depending on the source-preparation techniques employed. A plot of these results can be used, with caution, to estimate the self-absorption of a nuclide that decays by $\beta$ emission only and which is therefore not amenable to the coincidence technique.

In an attempt to overcome these objections we have investigated the possibility of combining with the $\beta$ activity a suitable $\beta\gamma$ nuclide. It is preferable to incorporate both activities into the same molecular structure in order to avoid any differential effects in the crystallization process as the source is dried. The overall efficiency of the $\beta$ detector for such a source of mixed activities is measured with respect to the $\beta\gamma$ nuclide using the $4\pi \beta\gamma$ coincidence technique. This efficiency is very largely determined by the self-absorption in the source material, and is also a measure of the efficiency of the $\beta$ detector with respect to the $\beta$ activity. Obviously it is advantageous to choose a $\beta\gamma$ nuclide whose $\beta$ end-point energy is similar to that of the $\beta$ nuclide. However, no two $\beta$ spectra are exactly alike and a correction must be made to allow for any difference. This may be made in the following manner. A number of sources of mixed $\beta$ and $\beta\gamma$ activities are prepared in which the amount of inactive carrier, and hence self-absorption, is varied. The $\beta$ detector efficiency is measured with respect to the $\beta\gamma$ nuclide for each source, and if this is a short-lived activity it is allowed to decay to a negligible level. The counting rate of the $\beta$ only activity is then measured in the same detector and this rate is then plotted against the corresponding efficiency for the $\beta\gamma$ nuclide. This plot yields over a considerable efficiency range a straight line which may be extrapolated to 100% efficiency to obtain the absolute counting rate of the $\beta$ only activity. Br$^{82}$ is suggested as one convenient $\beta\gamma$ nuclide. The method can be extended to the use of long-lived nuclides such as Co$^{60}$ thus giving a greater range of $\beta$ end-point energies. In this case the contribution of the $\beta\gamma$ nuclide to the total $\beta$ counting rate is determined from the efficiency and the amount of the $\beta\gamma$
activity added. This contribution is then subtracted to give the β counting rate due to the β activity.

It will be shown that the linear extrapolation is to be expected on theoretical grounds by considering various models for the self-absorption process. Assuming theoretical β spectral shapes the efficiency of one nuclide has been computed as a function of that of another. Although none of the models considered exactly corresponds to the experimental results described below they all indicate the presence of a linear relationship near 100% efficiency. The one exception found to date is the special case in which one nuclide of a pair is a positron emitter.

The extrapolation technique has been verified experimentally for a number of pairs of suitable β-γ nuclides in which the efficiency of the β detector can be measured with respect to both activities. Experimental results will be shown and compared with the theoretical models. In addition a sample of $^{35}$S has been standardized by this method using $^{60}$Co$^{35}$O$_4$ and Lu$_{177}^{177}(S^{36}O_4)_3$ and the results are in good agreement with those obtained by differential gas counting of $^{36}$S and by 4πβ counting using a self-absorption correction taken from the above mentioned curve.

Experience suggests that provided a β-γ nuclide can be found whose β endpoint energy is within a factor of roughly two of that of the β emitter the latter can be standardized absolutely to an accuracy of 1 to 3%.

Mesure absolue des nuclides ne se désintégrant que par émission β

Lorsque l'on détermine les taux absolus de désintégration de sources en utilisant la méthode du compteur 4πβ, il est difficile de mesurer ou même d'estimer avec quelque certitude la correction d'autoabsorption. Les résultats confirment la thèse selon laquelle, en diluant progressivement une solution mère jusqu'à ce que l'activité soit constante, on réduit seulement l'autoabsorption et on ne la supprime pas complètement. De plus, connaissant une série de valeurs de l'activité apparente correspondant à des valeurs de la densité superficielle moyenne de la source, il serait imprudent d'extrapoler.

Comme première approche du problème, les auteurs ont mesuré, par la méthode des coïncidences 4πβ-γ, l'autoabsorption en fonction de l'énergie β limite pour une densité superficielle moyenne de la source constante (2 µg/cm$^2$) et pour plusieurs méthodes de préparation des sources. L'autoabsorption variait, selon la méthode de préparation employée, de quelques dixièmes pour cent pour des énergies β limites de 1,35 MeV, à 20—40% pour 0,086 MeV. On peut utiliser avec précaution une représentation graphique de ces résultats pour évaluer l'autoabsorption d'un nuclide qui ne se désintègre que par émission β, et auquel la méthode des coïncidences ne convient pas.

Pour tenir compte de ces objections, les auteurs ont étudié la possibilité de combiner l'activité β avec un nuclide β-γ approprié. Il est préférable de faire en sorte que les deux activités soient contenues dans la même structure moléculaire afin d'éviter que des effets différentiels ne se produisent pendant la cristallisation au cours du séchage de la source. On mesure le rendement global du détecteur β pour une telle source d'activité mixte par rapport au nuclide β-γ en recourant à la méthode des coïncidences 4π β-γ. Ce rendement est déterminé surtout par l'autoabsorption de la matière brute et constitue aussi une mesure du rendement du détecteur β par rapport à l'activité β. Il est manifestement avantageux de choisir un nuclide β-γ dont l'énergie β limite est voisine de celle
du nuclide $\beta$. Cependant, il n'y a pas deux spectres $\beta$ identiques et il faut appliquer un facteur de correction pour tenir compte des différences. On peut procéder de la façon suivante: on prépare un certain nombre de sources d'activité mixte $\beta$ et $\beta-\gamma$ dans lesquelles on fait varier la quantité d' entraîneur inactif — et, de ce fait, l' autoabsorption. Dans chaque cas, on mesure le rendement du détecteur $\beta$ par rapport au nuclide $\beta-\gamma$; s'il s'agit d'une activité de courte période, on laisse le nuclide se désintégrer jusqu'à ce que l'activité devienne négligeable. Ensuite, on mesure dans le même détecteur le taux de comptage de l'activité $\beta$ seulement et on en fait la représentation graphique en fonction du rendement correspondant pour le nuclide $\beta-\gamma$. Pour un grand nombre de valeurs du rendement, la représentation donne une droite, ce qui permet d'extrapoler afin d'obtenir pour un rendement de 100% le taux de comptage absolu de l' activité $\beta$ seulement. Comme nuclide $\beta-\gamma$ approprié, les auteurs proposent le Br$^{82}$. On peut étendre l'emploi de cette méthode au cas de nuclides de longue période, tels que le Co$^{60}$, ce qui donne une gamme élargie d'énergies $\beta$ limites. Dans ce cas, l'apport du nuclide $\beta-\gamma$ au taux global de comptage $\beta$ est déterminé d'après le rendement et la quantité d'activité $\beta-\gamma$ ajoutée. On soustrait ensuite cet apport pour obtenir le taux de comptage $\beta$ résultant de l'activité $\beta$.

Les auteurs montrent que, pour des raisons théoriques, on peut s'attendre à obtenir une ligne droite, en considérant divers modèles du processus d'autoabsorption. En admettant les formes théoriques des spectres $\beta$, on a pu calculer le rendement d'un nuclide en fonction du rendement d'un autre. Bien qu'aucun des modèles considérés ne corresponde exactement aux résultats expérimentaux exposés ci-après, ils indiquent tout l'existence d'un rapport linéaire pour les valeurs du rendement voisines de 100%. La seule exception que l'on ait trouvée jusqu'ici est le cas particulier où l'un des nuclides pris deux à deux est un émetteur de positrons.

La méthode d'extrapolation a été contrôlée expérimentalement pour plusieurs couples de nuclides $\beta-\gamma$ pour lesquels le rendement du détecteur $\beta-\gamma$ peut être mesuré pour les deux activités. Les auteurs donnent les résultats obtenus expérimentalement et les comparent aux modèles théoriques. En outre, on a établi par cette méthode un échantillon de S$^{35}$ en utilisant le Co$^{60}$S$^{35}$O$^4$ et du Lu$^{177}$(S$^{35}$O$^4$) pour lesquels on a pu calculer un rapport linéaire pour les valeurs du rendement voisines de 100%. La seule exception que l'on ait trouvé jusqu'ici est le cas particulier où l'un des nuclides pris deux à deux est un émetteur de positrons.

L'expérience semble indiquer qu'à condition de pouvoir trouver un nuclide $\beta-\gamma$ dont l'énergie $\beta$ limite est comprise entre la moitié et le double de celle de l'émetteur $\beta$, il est possible de faire l'étalonnage absolu de ce dernier avec une précision de 1 à 3%.

**Абсолютный счет распада изотопов только при помощи $\beta$ излучения**

В определении скоростей абсолютного распада источников, когда используется метод $4\pi \beta$ счета, трудно измерить или даже подсчитать поправку на самопоглощение с любой степенью точности. Результаты подтверждают точку зрения о том, что метод прогрессивного разжижения запаса раствора до получения постоянной активности лишь сводит самопоглощение к минимуму, но не устраняет его. Кроме того, не рекомендуется применение любого приема экстраполяции, основанного на графике кажу-
щественной активности по отношению к средней поверхностной плотности источника.

Поэтому в качестве первого подхода к решению проблемы, использовав метод 4 π β-γ счета совпадений, мы измерили самопоглощение как функцию β-излучений и границной энергии для поверхностной плотности постоянного среднего источника 2 мкГ/см² и для ряда приемов по подготовке источника. Самопоглощение колебалось от нескольких десятых процента при β излучении средней энергии в 1,39 MeV до 20—40% при 0,086 MeV в зависимости от применяемого метода подготовки источника. График этих результатов может быть с осторожностью использован для подсчета самопоглощения изотопа, который распадается лишь при воздействии β излучения и на который поэтому не распространяется метод счета совпадений.

Пытаясь преодолеть эти трудности, мы исследовали возможность соединения изотопа, обладающего β-γ активностью с соответствующим изотопом, обладающим β активностью. Предпочтительно соединять обе активности в одну молекулярную структуру с тем, чтобы избежать любых дифференциальных эффектов в процессе кристаллизации при сушке источника. Полная эффективность β детектора на такой источник смешанной активности измеряется в отношении β-γ изотопа методом 4 π β-γ счета совпадений. Эта эффективность в значительной степени определяется самопоглощением в исходном материале и является критерием эффективности β детектора по отношению к β активности. Очевидно более полезным является выбор изотопа с β-γ излучением, β излучения и границная энергия которого подобны таковым, имеющимся у изотопа с β излучением. Однако вы не найдете двух сходных β спектров, поэтому следует делать поправку на разницу. Это можно осуществить следующим образом. Приготовляется ряд источников со смешанной β и β-γ активностью, в которых различается количество неактивного носителя, а следовательно и самопоглощение, эффективность β детекторов измеряется в отношении изотопа с β-γ излучением на каждый источник и если это короткоживущая активность, то она распадается до незначительного уровня. Скорость счета лишь β активности тогда измеряется в том же детекторе и наносится на график против соответствующей активности для изотопа с β-γ излучением. Этот график представляет собой через значительный диапазон эффективности прямую линию, которую можно экстраполировать до 100%-й эффективности с тем, чтобы получить абсолютную счетную скорость лишь β активности. В качестве одного из подходящих изотопов с β-γ активностью рекомендуется Br82. Данный метод может использоваться долгоживущие изотопы, такие как Co-60, придавая этим большей диапазон β излучению и границной энергии. В этом случае составляющая β-γ активности изотопа в общей скорости отсчета β активности определяется в зависимости от эффективности и количества добавленной β-γ активности. Эта составляющая затем вычитается с тем, чтобы определить скорость отсчета β активности при помощи β активности.

Можно показать, что линейную экстраполяцию можно ожидать на теоретической основе путем изучения различных видов процесса, связанного с самопоглощением. Учитывая теоретические β спектральные формы, эффективность одного изотопа исчислялась в качестве функции другого. Несмотря на то, что ни один из рассмотренных видов точно не соответствовал описанным ниже экспериментальным результатам, все они указывали...
вают на наличие линейной связи почти 100%-ой эффективности. На сегодняшний день обнаружено лишь одно исключение — это особый случай, в котором один из парных изотопов является излучателем позитронов.

Приемы экстраполяции проверялись в ходе экспериментов на ряде пар соответствующих изотопов с β-γ активностью, в которых эффективность β детектора может измеряться в отношении обеих активностей. Будут приведены результаты экспериментов и будет дано их сравнение с теоретическими видами. В дополнение к этому с помощью данного метода стандартизировался образец S35, используя Co60S35O4 и Lu2177 (S35O4), и результаты стандартизации находятся в полном соответствии с теми результатами, которые были получены при дифференциальном газовом счете S35O2 и при 4π β счете с учетом поправки на самопоглощение, взятой из вышеупомянутой кривой.

Recuento absoluto de los núcleidos que se desintegran por emisión β exclusivamente

Para determinar las velocidades absolutas de desintegración de fuentes radiactivas por el método de recuento β 4π, es difícil medir e incluso evaluar con cierto grado de aproximación la corrección por autoabsorción. Los resultados corroboran la hipótesis de que por dilución progresiva de una solución madre hasta alcanzar una actividad constante, sólo se logra reducir la autoabsorción, pero no anularla. Por otra parte, el empleo de procedimientos de extrapolación basados en la representación gráfica de la actividad aparente en función de la densidad superficial media de la fuente, no es recomendable.

Para abordar el problema, los autores, utilizando el método de coincidencias β-γ 4π, han determinado la autoabsorción en función de la energía límite de los rayos β para una densidad superficial media de la fuente constante e igual a 2 μg/cm² y para diferentes procedimientos de preparación de las fuentes. De acuerdo con el procedimiento utilizado para la preparación de la fuente, la autoabsorción varió de unas décimas de por ciento, para energías β límite del orden de 1,39 MeV, hasta 20—40%, aproximadamente, para energías del orden de 0,086 MeV. Con ciertas reservas puede usarse una representación gráfica de estos resultados para calcular la autoabsorción de un núcleo que se desintegra emitiendo exclusivamente rayos β y al que, por esta razón, no puede aplicarse el método de coincidencias.

Con el propósito de superar estos inconvenientes, los autores han estudiado la posibilidad de asociar a los emisores β un núcleo β-γ adecuado. Es conveniente incorporar ambos núcleidos en la misma estructura molecular a fin de prevenir efectos diferenciales en el proceso de cristalización al secar la fuente. La eficiencia global del detector β para esta fuente de actividades mixtas se mide en función del núcleo β-γ, empleando el método de coincidencias β-γ 4π. Dicha eficiencia depende en gran medida de la autoabsorción que se produzca en la sustancia de la fuente, y es también un índice de la eficacia del detector β respecto de la actividad β. Es evidente que conviene elegir un núcleo β-γ cuya energía β límite sea análoga a la del emisor β. Sin embargo, como no existen dos espectros β exactamente iguales, es preciso efectuar correcciones a fin de tener en cuenta las diferencias. Para ello, puede procederse de la siguiente manera: se preparan varias fuentes de núcleos β y β-γ mezclados, en las que varía la cantidad de portador inactivo y, por tanto, la autoabsorción. Se mide la eficiencia del detector β en función del núcleido β-γ de cada fuente, y si éste
es de período corto, se espera hasta que su actividad alcance un valor despreciable. A continuación se mide la velocidad de recuento correspondiente a la actividad β, exclusivamente, con el mismo detector, y dicha velocidad se representa gráficamente en función de la eficiencia referida al núcleo β-γ. El gráfico así obtenido, constituye para una amplia gama de eficiencias una recta que puede extrapolarse hasta una eficiencia del 100%, lo que permite determinar la velocidad de recuento absoluta correspondiente a la actividad β exclusivamente. Como núcleo β-γ se recomienda el Br82. Este método también puede utilizarse con núcleos de período largo como el Co60, con lo que se obtiene una mayor variedad de energías β límite. En este caso se determina la contribución del núcleo β-γ a la velocidad total de recuento β sobre la base de la eficiencia del contador y de la cantidad de núcleo β-γ añadida. Después se resta esa contribución para hallar la velocidad de recuento β que corresponde efectivamente al emisor β.

Los autores demuestran, analizando diferentes modelos para el proceso de autoabsorción, que por razones teóricas la extrapolación lineal es probablemente la correcta. Suponiendo que la configuración de los espectros β se ajusta a la teórica, se ha calculado el rendimiento de un núcleo en función del rendimiento de otro. Si bien ninguno de los modelos estudiados corresponde exactamente a los resultados experimentales expuestos, todos ellos confirman la existencia de una relación lineal cuando el rendimiento se acerca al 100%. La única excepción encontrada hasta hoy es el caso especial en que uno de los dos núcleos del par es emisor de positrones.

La utilidad del método de extrapolación ha sido verificada experimentalmente para una serie de pares de núcleos β-γ con los que puede medirse la eficiencia del detector β en función de las dos actividades. Los autores dan a conocer los resultados experimentales obtenidos y los comparan con los modelos teóricos. Además, han calibrado una muestra de S35 por este método, utilizando Co60S35O4 y Lu177(S35O4); los resultados concuerdan satisfactoriamente con los obtenidos por recu otendiferencial del S35O4 gaseoso y por el método β 4π, siempre que se aplique la corrección por autoabsorción determinada mediante la curva antes mencionada.

Los experimentos parecen confirmar que siempre que se disponga de un núcleo β-γ cuya energía β límite no sea superior al doble de la del emisor β, este último puede calibrarse en valor absoluto con una precisión del 1 al 3%.

The greatest uncertainty in 4πβ counting using deposited sources usually arises from the absorption of the β particles in the source material itself, or the self-absorption as it is generally called. The difficulties of making reliable self-absorption corrections are well known and often it has been customary not to make such corrections but to quote a disintegration rate with a sufficiently large error to take care of self-absorption. If you recall the comparison of the results obtained for Au198 by the 4πβ and 4πβ-γ coincidence methods (Paper 1. 1), very good agreement was obtained by different laboratories using the coincidence method, but there was the usual 1—2% spread in the results obtained by straight 4πβ counting. This spread is probably due to variation in source preparation techniques and hence to variation in self-absorption. The problem before us therefore is to improve the accuracy of the standardization of those nuclides which do not have a γ-ray in their decay
scheme, the so-called pure β emitters. Such improvement must necessarily be made by paying particular attention to the question of self-absorption.

At Chalk River considerable attention has been given to this problem and I would like now to describe some studies which we have made in this respect. Some of this work has recently appeared in the literature (1), other parts will be published shortly. In our examination of the problem we have used conventional source-preparation techniques which may produce local agglomerates of considerable size. The term "mean superficial density" in the present paper should therefore be understood as the average over a considerable range of source thickness in contrast to the more uniform distributions which Professor Yaffe used in his approach to this problem (Paper II. 2. 2.). The present studies all hinge on the fact that in the coincidence method the overall efficiency of the β detector can be readily obtained from the ratio of the coincidence counting rate to the γ channel counting rate. If pulse-height selection is used in the γ channel it follows that the efficiency measured in this way is that of the β group associated with the γ-ray so selected. Thus, for example, it is possible to measure the efficiency of an inner β-ray group in the presence of a ground state β transition. If the β detector is a conventional 4π gas

![Figure 1](image)

The self-absorption as a function of the mean superficial density for Nb$^{95}$, the 680 keV transition of Rb$^{86}$, and Au$^{198}$. The inset shows the variation of self-absorption at low mean superficial densities for Nb$^{95}$.
counter then the overall efficiency is determined primarily by self-absorption; the source-mount absorption and any intrinsic inefficiency in the counter-amplifier system being considerably less \(^{(1)}\). The source-mount absorption can be readily determined by an extrapolation technique, while because of its very small value reliable estimates of the correction due to the threshold detection limit of the counter-amplifier system can be made. Thus from a measurement of the overall efficiency the self-absorption of a source may be readily determined. A brief description of the \(4\pi\) \(\beta-\gamma\) coincidence unit that was used in these studies was given in Paper I. 1 and has been fully described in the literature \(^{(2)}\).

As a first approach to the problem we have measured the self-absorption as a function of the mean superficial density of the source material for \(\text{Nb}^{95}\), the 680 keV branch of \(\text{Rb}^{88}\), and \(\text{Au}^{198}\) using conventional source preparation techniques \(^{(4)}\). The results are shown in figure 1. The shaded area in each case gives an indication of the scatter of the experimental points and shows that as the source thickness increases this scatter also increases. The knee is a characteristic feature of these curves and may perhaps be associated with the transition from isolated crystals to a continuous distribution of active material.

![Figure 2](image-url)

Figure 2
The self-absorption as a function of \(\beta\) end-point energies for sources of 2 \(\mu\)g/cm\(^2\) mean superficial density.
over the area of the source. At very low mean superficial densities the curve shows a further turning point and enters a region in which the self-absorption is approximately independent of mean source thickness (see inset in figure 1 for Nb\(^{95}\)). This is undoubtedly due to the fact that further dilution of the active solution does not produce smaller crystals but merely increases the distance between them. It is clear from these results that any attempt to extrapolate a curve of counting rate against mean superficial density to obtain an absolute disintegration rate using conventional source preparation techniques is not going to yield a very accurate answer owing to the large scattering in the data.

In a further study we have measured the self-absorption as a function of \(\beta\)-ray end-point energy for a constant mean source superficial density of 2 \(\mu g/cm^2\) (1). Some of these results are shown in figure 2 where the self-absorption is plotted as a function of the end-point energy. Curve A corresponds to sources prepared by the simple evaporation technique, that is drying under a heat lamp, while curve B represents sources prepared by the colloidal silica treatment. All sources are 2 \(\mu g/cm^2\) mean superficial density. The errors represent the standard deviation of about 5 sources and again show that the variation in self-absorption becomes larger as the self-absorption itself becomes greater. Sources treated with a wetting agent such as insulin yielded results which were very similar to curve B and one readily see that a considerable improvement is obtained by the use of either colloidal silica or insulin, particularly for low \(\beta\) end-point energies. There is one notable deviation from these smooth curves and that is the value for the positron branch of Cu\(^{64}\). This is due to the fact that, owing to the coulomb repulsion effect, there are very few low energy particles in a positron spectrum.

The results of figure 2 can be approximated by the following empirical relation:

\[
\text{Self-absorption} \approx \frac{k}{\bar{W}_{\beta}} \quad \ldots \ldots \ldots \ldots (1)
\]

where \(\bar{W}_{\beta}\) is the average \(\beta\) particle energy and \(k\) is a constant dependent on the source thickness. Figure 3 shows the self-absorption data of figure 2 plotted against the inverse average \(\beta\) particle energy, and clearly there is a roughly linear relationship between these quantities. The full lines are a least squares fit to the data assuming equation (1) to be correct.

Provided that the source-preparation techniques and mean source superficial densities are the same as those employed in these studies, curves such as those of figures 2 or 3 may be used with caution to estimate self-absorption corrections. It is necessary to emphasize the 'with caution' since the use of these curves in this manner is always open to the two objections that the shape of the beta spectrum and the form of the crystalline deposit may vary from nuclide to nuclide. The case of Cu\(^{64}\) is an extreme example of this variation but amply demonstrates that due care must be taken. The representations shown in figure 3 clearly indicate the error that may be associated with such a procedure.

In order to avoid these difficulties of spectral shapes and differing forms of crystalline deposits we have investigated the use of a suitable \(\beta\)-\(\gamma\) emitting nuclide added directly to the pure \(\beta\) activity so that both nuclides are contained in the same chemical compound. Since such a \(\beta\)-\(\gamma\) activity is used to determine,
or trace, the efficiency of the pure $\beta$ emitter we call this method the tracer technique. Ideally one would like to have the $\beta$ spectral shapes of the pure $\beta$ emitter and the tracer identical. If in addition to this the tracer nuclide decayed with a much shorter half-life than that of the pure $\beta$ emitter it would then be possible to make a measurement of the overall efficiency of the $\beta$ detector with respect to the $\beta$-$\gamma$ nuclide, allow this activity to decay, and then apply this efficiency to the straight $4\pi$ count due to the pure $\beta$ emitter. In practice, as we have already noted, the shapes of $\beta$ spectra vary considerably from nuclide to nuclide and hence these ideal conditions cannot be attained. However the method can be made quite practicable in the following way: A series of sources containing both the pure $\beta$ activity and a ‘spike’ of a suitable tracer activity is prepared, each source having a different amount of inactive
carrier added, and hence a different mean superficial density. However the actual values of these densities need not be known. The overall efficiency of the $\beta$ detector is measured with respect to the tracer activity which is then allowed to decay. The observed counting rate in the $4\pi$ counter due to the remaining pure $\beta$ activity in each source is plotted as a function of the corresponding efficiency measured with respect to the tracer nuclide. This yields a straight line which may be extrapolated to 100\% efficiency to obtain the absolute disintegration rate of the pure $\beta$ emitter. I will discuss the justification for this linear extrapolation in detail in a minute, but first, just to orient our thinking, I would like to suggest as one possible candidate for a tracer activity, the nuclide Br$^{82}$. It has a conveniently short half-life, about 36 hours, which enables the whole measurement to be completed within 2 weeks, and essentially all disintegrations take place via one $\beta$-ray group having an end-point energy of 444 keV. The $\beta$ emission is followed by a large number of $\gamma$-rays which is a further advantage since it effectively increases the efficiency of the $\gamma$ detector and hence only a small quantity of the tracer need be added.

There is an obvious extension of the method in which long lived $\beta$-$\gamma$ nuclides can be used as tracers. The contribution of the tracer nuclide to the total $\beta$ counting rate in a mixed source is determined from the efficiency measurement and the amount of the $\beta$-$\gamma$ activity added, which, in this case, must be known. This contribution is then subtracted from the total $\beta$ counting rate to give the counting rate due to the $\beta$ activity. Thus, with this modification a much wider choice of tracers is available giving a greater range of $\beta$ end-point energies.

We must now examine the justification for the linear extrapolation which is of course basic to the whole technique. The evidence falls into two parts, theoretical calculations and experimental proof. For this part of the discussion it is convenient to define the term ‘escape probability’, $P(W)$, as the probability that an electron of energy $W$ will escape from a mass of active material, which in this case will be a $4\pi$ $\beta$ source. Since it has been shown that, compared to self-absorption, the film absorption is small (1) we include the effects of the latter in this escape probability. If the form of this escape probability as a function of energy is known then, assuming a theoretical $\beta$ spectral shape, numerical integrations can be made to calculate the efficiency as a function of some parameter contained in the escape probability. If, for example, we take a typical $\beta$ spectrum shown at the top of figure 4 and assume for the moment that the correct form of the escape probability is the step function shown in figure 4, centre, then the spectrum of $\beta$ particles actually entering the gas of the counter is given by the product of these two functions and is shown at the bottom of figure 4. Thus the efficiency of the beta detector is simply the ratio of the shaded area to the total area under the $\beta$ spectrum, and can be calculated as a function of $W_t$, the cut-off energy. This form of the escape probability is obviously not exact but in point of fact it does give fairly good results. More realistic guesses at the escape probability are the exponential form

$$P(W) = 1 - e^{-\mu W}$$

and gaussian form

$$P(W) = 1 - e^{-\mu^2 W^2}.$$
In both of these expressions the variable parameter is of course \( \mu \). In addition two further functions for the escape probability were derived. LANE and ZAFFARANO (3) have studied the penetration of thin foils by a well collimated beam of monoenergetic electrons in the energy range below 40 keV which is the range of interest to the present problem. Their experimental results are expressed as a probability of penetration as a function of energy for several thicknesses of foil and we have found that their results can be fitted fairly well by an equation of the form

\[
P_i(W) = 1 - e^{-\frac{W}{\mu A}}.
\]

By comparing their results as a function of foil thickness we find that \( \mu A \approx \text{constant} \approx 2 \), and hence there is in effect just one variable parameter in this equation. Thus it is possible to use some experimental results to calculate the escape probability of electrons from a radioactive source. We assume that the activity is uniformly distributed in an infinite slab of material or into well separated spheres for which the necessary geometrical integrations can be made and we are indebted to Dr. Kennedy for this part of the work. For \( \mu A = K \) the final equations are:
\[ P(W) = \frac{W}{2t} e^{K E_1(K)} \quad \text{for } W < t \]

\[ = 1 - \frac{t}{2W} \left[ 1 + \frac{1}{K} - \frac{2 e^K}{K} E_3 \left( \frac{KW}{t} \right) \right] \quad \text{for } W > t \]

\[ P(W) = \frac{3}{4} \frac{W}{R} \left[ 1 - \frac{1}{12} \frac{W^2}{R^2} - e^K E_2(K) + \frac{1}{4} \frac{W^2}{R^2} e^K E_4(K) \right] \quad \text{for } W < 2R \]

\[ = 1 - \frac{3 e^K}{2} \left[ E_2 \left( \frac{KW}{2R} \right) - E_4 \left( \frac{KW}{2R} \right) \right] \quad \text{for } W > 2R \]

where \( t \) is the thickness of the slab, \( R \) the radius of the spheres and \( E_1, E_2, \) etc. are the functions of CASE et al. (4).

These above five expressions for \( P(W) \) have been used to calculate efficiencies as a function of the variable parameter for a number of nuclides using theoretical shapes for the \( \beta \) spectra (5). The calculations for the first expression were done by hand while the remainder were programmed on the Chalk River Datatron computer. Some typical results obtained from these calculations are shown in figure 5. The efficiency of one nuclide is shown as a function of the efficiency of another, the variable parameter being the same for both nuclides at any one point on the curve. The points, which are connected by the dashed lines, are experimentally determined values and will be discussed in a minute. The results for several pairs of nuclides are shown, the \( \beta \) transition being specified if there are two or more \( \beta \) branches in the same nuclide. The solid curves are calculated using the expressions for the escape probability as shown. In most cases they do not show very striking differences between one another and in some cases, such as \( \text{Ce}^{141} (440 \text{ keV}) - \text{Br}^{82} \), it is impossible to distinguish between some curves. However all five forms for the escape probability predict a closely linear relationship between the efficiencies of pairs of nuclides. There is one deviation from this generalization, namely, the curve for the efficiency of \( \text{Na}^{24} \) against that of \( \text{Na}^{22} \) which shows a slight knee close to 100\% efficiency. This prediction is also borne out by the experimental results, and arises from the characteristic spectral shape of a positron emitter which as we have already noted has very few low energy particles.

The linearity relationship has been verified experimentally by taking pairs of suitable \( \beta-\gamma \) nuclides. For example, we may take the case of \( \text{Ce}^{141} \) and \( \text{Br}^{82} \). \( \text{Ce}^{141} \) has two \( \beta \) groups, one of which has an end-point energy of 440 keV and is followed by a \( \gamma \)-ray of 149 keV, while the other group is the transition to the ground state of the daughter. The half-life of this nuclide is 35 days. The two activities were made up in the form of \( \text{CeBr}_3 \) and the overall efficiency of the \( \beta \) detector with respect to the \( \text{Br}^{82} \) activity was measured by gating the \( \gamma \) channel well above the 149 keV \( \gamma \)-ray of \( \text{Ce}^{141} \). The \( \text{Br}^{82} \) activity was allowed to decay and the efficiency of the inner \( \beta \) group of \( \text{Ce}^{141} \) was then measured by gating on the 149 keV \( \gamma \)-ray. One problem which has arisen from these studies is the question of source stability. It is of course essential that the physical form of a source remain unchanged between measurements, but unfortunately some bromides tend to be deliquescent. To minimize this effect therefore all sources were stored when not in use in a dessicator and while this has helped considerably I think that the effect is perhaps still present to a small extent as evidenced by
Figure 5
Efficiency functions for various pairs of β groups. The full curves are derived theoretically while the points are experimental and are connected by broken lines.

the slight scatter of the experimental points which seems to be somewhat greater than might be expected. We are currently investigating the use of bromates instead of bromides and the hope is that sources of these compounds will show better stability.

The results for the Br$^{82}$ and the 271 keV transition in Fe$^{59}$ for Br$^{82}$ and the 178 keV transition in Lu$^{177}$, for Na$^{24}$ and Br$^{82}$ and for Na$^{24}$ and Na$^{22}$ were obtained in a similar way. In each case the two activities were contained in the same molecular structure. The two iron β groups are of course contained in the same nuclide. The efficiency measurement for the 271 keV β group of course merely requires that the γ channel be gated on the highest energy peak in the γ-ray spectrum of Fe$^{59}$. The efficiency measurement with respect to the 460 keV β transition was made by placing a narrow gate across the 1.29 MeV photo
peak, which fortunately lies in the valley of the 1.33 MeV γ-ray distribution, and, using standard peak shapes, a correction made for those pulses which are associated with the 271 keV β group. This correction amounted to less than 10%.

The linear relationship is well demonstrated in the top four examples of figure 5. The case of Na$^{24}$ and Br$^{82}$ is an extreme one since we are comparing a 1.4 MeV end-point β emitter with one having only 444 keV end-point energy — a factor of more than 3 smaller. Such a large difference would be avoided in practice if possible. However, the results for Na$^{24}$ and Br$^{82}$ still exhibit a straight line with about the same slope as predicted although the line fails to extrapolate to 100% by a very small amount. In view of the difference in end-point energies this is not unreasonable. The comparison of the 178 keV branch in Lu$^{177}$ with Br$^{82}$ is another extreme example — here the ratio of end-point energies is about 2.5, but again, the points fall fairly well on a straight line. Experimentally, this was a difficult comparison to make since the 178 keV β branch is only a few per cent abundant in Lu$^{177}$ and two γ-rays must be summed in order to ensure that only the efficiency of this branch is measured. As has been mentioned above the experimental curve for Na$^{24}$ and Br$^{82}$ shows a similar shape to some of the theoretical curves but deviates from them by a few tenths of one per cent.

These experimental results confirm the assumption of a linear relationship between efficiencies for those cases considered. The results suggest that in any practical application of this technique the end-point energies of the two nuclides should for preference be within a factor of two of each other. It is obvious from these results that no one of the five escape probabilities can exactly describe all the observed data. One can note however one or two generalizations. For example, the slab type calculations deviate considerably from the experimental points, probably because of the poor assumption of an infinite slab for thin $4\pi$ sources. On the other hand the sphere and gaussian type calculations give results which agree very closely with one another and are in fair agreement with the experimental data. We have therefore used the gaussian form to calculate the efficiency as a function of the variable parameter $1/\mu$ for a large number of nuclides, including those which were used to set up the self-absorption curves of figure 2. The parameter $1/\mu$ is of course the energy of an electron which has a 63% chance of escape and if the gaussian form for the escape probability can be used to describe the physical situation in all $4\pi$ counting sources, then $1/\mu$ should have the same value for all these nuclides provided that the superficial density is the same for all sources. Table I shows a list of these nuclides, their measured self-absorptions ($\tau$), and the calculated values of $1/\mu$ in kilovolts. While the self-absorption varies over quite a considerable range, from almost zero up to 40%, $1/\mu$ is practically constant within the errors. I should perhaps emphasize that all these results depend on theoretical β spectral shapes, particularly in the region below, say, 50 kilovolts which has not been investigated by β-ray spectrometry too well. The fact that reasonable agreement is obtained may indicate that at least there are no sharp deviations in the actual spectral shapes of these nuclides from the theoretical predictions.

Additional experimental evidence in favour of this technique has been obtained from the standardization of S$^{35}$ using both Co$^{60}$ and Lu$^{177}$ as the tracer nuclides, the activities being in the form of sulphates, which incidentally are not deliquescent. Figure 6 compares those results obtained by the tracer tech-
TABLE I

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>End-Point Energy of Group, MeV</th>
<th>Evaporated Sources</th>
<th>Colloidal Silica Treated Sources</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Self-Absorption</td>
<td>μ⁻¹ keV</td>
</tr>
<tr>
<td>I⁰</td>
<td>0.086</td>
<td>39 ± 6</td>
<td>16.2 ± 2.8</td>
</tr>
<tr>
<td>I⁰</td>
<td>0.16</td>
<td>20.3 ± 2.1</td>
<td>15.8 ± 1.8</td>
</tr>
<tr>
<td>H⁰</td>
<td>0.21</td>
<td>8.7 ± 1.5</td>
<td>14.5 ± 2.0</td>
</tr>
<tr>
<td>C⁰</td>
<td>0.31</td>
<td>5.3 ± 1.7</td>
<td>11.6 ± 3.7</td>
</tr>
<tr>
<td>Sc⁶⁶</td>
<td>0.36</td>
<td>0.2 ± 0.2</td>
<td>24 ± 6</td>
</tr>
<tr>
<td>Cu⁸⁴(β⁺)</td>
<td>0.66</td>
<td>2.7 ± 1.1</td>
<td>13.0 ± 5.5</td>
</tr>
<tr>
<td>Rb⁸⁶</td>
<td>0.68</td>
<td>1.6 ± 0.6</td>
<td>10.6 ± 3.8</td>
</tr>
<tr>
<td>Au¹⁹⁸</td>
<td>0.96</td>
<td>1.0 ± 0.5</td>
<td>28 ± 13</td>
</tr>
</tbody>
</table>

The tracer technique has also been used to determine the absolute disintegration rate of P³² in the recent (April 1959) international intercomparison of this nuclide. In this case Na²⁴ was combined with P³² in the form of NaH₂PO₄ and the results are shown in figure 7 A. A summary of this inter-
MEAN SUPERFICIAL DENSITY mg/cm²

Figure 7

A. The $^{32}P$ counting rate as a function of the efficiency of Na$^{24}$ tracer.

B. The $^{32}P$ counting rate as a function of source mean superficial density.

comparison has been circulated to the laboratories taking part (6). For comparison figure 7 B shows the $^{32}P$ counting rate as a function of the source mean superficial density for the same sources as were used in the tracer experiment.

Finally, we would like to suggest that this technique may have a particular application to those coincidence systems which have a solid crystal or liquid scintillation detector for the 4 $\pi$ counter. In these arrangements essentially all the energy of the $\beta$ particles is expended within the detector and pulse-height selection is therefore possible. Thus in principal the step function form for the escape probability can be imposed artificially by the introduction of a discriminator. This discriminator should be set well above any real self-absorption effects or noise pulses. Thus the escape probability — this is of course a misnomer in this application, is known exactly and it is a fairly easy matter to compute efficiency ratios for it. In actual practice, because of the finite resolution of the detector, the cut-off will not be absolutely sharp but a correction can be made for this. Further, only measurements on one mixed source are required in principle to determine the disintegration rate of the pure $\beta$ emitter.
REFERENCES


A Quick Method of Measuring and Analyzing $\beta$ Emitters in Solution of Very Low-level Specific Activity

1. Description of a measuring instrument with a low-level background, for measuring radioactive materials in liquid form. Essentially the instrument consists of two flat counters, each with one hole, and one flat counter with two holes, mounted parallel to each other and enclosed in a casing of counters mounted in anti-coincidence with the inner counter.

2. Possible applications of the method:
   a) To measure much lower-level specific activities than with liquid counters, and particularly soft $\beta$ emitters;
   b) To detect radioactive impurities within a radionuclide (e. g. 2/10,000 of $\text{P}^{32}$ in $\text{S}^{35}$);
   c) For most radionuclides, to measure, without previous concentration, specific activities corresponding to the tolerance limits permitted at present.

Remarques sur une technique rapide de mesure et d’analyse d’émetteurs $\beta$ en solution de très faible activité spécifique

1. Description d’un dispositif de mesure à faible mouvement propre, destiné à la mesure de produits radioactifs sous forme liquide, et qui comprend essentiellement 2 compteurs plats à une fenêtre, et un compteur plat à deux fenêtres, disposés parallèlement et enfermés à l’intérieur d’une gaine de compteurs montés en anticoïncidence avec les premiers.

2. Possibilités et domaines d’utilisation de la méthode:
   a) permet de mesurer des activités spécifiques beaucoup plus faibles qu’avec les compteurs à liquides, particulièrement avec les émetteurs $\beta$ mous;
   b) permet de déceler des impuretés radioactives au sein d’un radioélément (ex. 2/10,000 de $\text{P}^{32}$ dans $\text{S}^{35}$);
   c) permet, pour la plupart des radioéléments, de mesurer, sans concentration préalable, des activités spécifiques égales respectivement aux limites de tolérance aujourd’hui admises.
2. Возможности и области использования метода:

а) этот метод позволяет измерять гораздо более слабую специфическую активность, чем это возможно с жидкотными счетчиками, в частности с излучателями не жестких β-частиц;

б) этот метод позволяет обнаруживать радиоактивные примеси внутри радиоэлемента (напр. 2/10 000 P<sup>32</sup> в S<sup>35</sup>);

в) этот метод позволяет для большинства радиоэлементов измерять без предварительной концентрации специфическую активность соответственно равную принятым в настоящее в настоящее время предельным допускам.

Observaciones sobre un método rápido de medición y análisis de emisores β en soluciones de muy baja actividad específica

1. Los autores describen un dispositivo de medición poco sensible a la actividad de fondo, destinado a medir la radiactividad de sustancias líquidas. El dispositivo consta fundamentalmente de dos contadores planos provistos de una ventana y de un contador plano con dos ventanas, disqueados paralelamente en el interior de una vaina de contadores conectados en anticoincidence con los primeros.

2. Además enumeran las posibilidades y campos de utilización del método que:

а) especialmente en el caso de los emisores β blandos, permite medir actividades específicas mucho más bajas que cuando se utilizan contadores para líquidos;

б) permite descubrir las sustancias radiactivas presentes como impurezas en determinados radioelementos (por ejemplo, 2/10 000 de P<sup>32</sup> en S<sup>35</sup>);

в) para la mayor parte de los radioelementos permite medir, sin necesidad de proceder a una concentración previa, actividades específicas de igual orden de magnitud que los límites de tolerancia actualmente admitidos.

Cette technique est essentiellement destinée à mesurer l'activité d'émetteurs β en solutions d'auSSI faible activité spécifique que possible, sans qu'il soit nécessaire de concentrer au préalable ces solutions ni de préparer des sources solides. Le but recherché est également de faire des mesures relativement rapides de façon à ce que cette technique puisse être utilisée pour des déterminations de routine.

Le compteur à liquide ne pouvait être retenu en raison de la notable absorption des β par les parois; le compteur à scintillateur liquide ne pouvait l'être non plus car on désirait une technique relativement indépendante de la composition chimique de la solution, en particulier des teneurs éventuelles d'entraineur. Pour atteindre un seuil de détection aussi bas que possible il est nécessaire de disposer d'un détecteur à mouvement propre peu élevé et à géométrie la plus grande possible.

Dispositif expérimental

1. Dé te c te u r — Il est constitué par deux compteurs GM plats à fenêtre, disposés face à face et montés en anticoïncidence avec une gaine de 28 compteurs de 80 cm de long. Les deux premiers compteurs sont logés dans
une boîte de bismuth, ceinturée par la gaine, elle-même entourée par une paroi de 20 cm de fer. Diamètre des fenêtres des compteurs: 50 mm. Mouvement propre pour l'ensemble des deux compteurs: 11 impulsions/minute. Ce mouvement propre résiduel est dû essentiellement au mica des fenêtres. Avec des compteurs à fenêtre de mylar il est raisonnable d'escompter un mouvement propre de 2—3 i/min. La figure 2 représente une variante — décrite plus loin — du dispositif, lequel dans cette première partie ne comprend que deux compteurs: C1 et C3, le compteur C1 occupant la position de C2.

2. Source radioactive — La solution radioactive est contenue dans une cuve cylindrique en plexiglass dont le fond est constitué par un film de Mylar (0,7 mg/cm²). Diamètre de la cuve: 60 mm. Cette cuve est placée entre les deux compteurs plats précités et de façon que la face libre du liquide et la paroi de mylar soient respectivement aussi près que possible des deux fenêtres; ainsi est-on assuré qu'une légère erreur de centrage est sans influence sur la mesure.

Il est important que la surface du liquide soit plane. On évite la formation d'un ménisque en essuyant la cuve, avant remplissage, avec un coton imbibé d'un agent mouillant. Des divers agents essayés, le Tween 20 (Polyoxy-éthylène-sorbitan monoléate) et le cétalon (bromure de cétyltriméthyl ammonium + acétone + éthanol) ont, dans l'ordre, donné les meilleurs résultats. Si l'on n'utilise pas de mouillant, la quantité de solution nécessaire pour être en "couche épaisse" au centre, pour S³⁵ et C¹⁴, est d'au moins 3 cm³. Avec mouillant, cette condition, nécessaire pour avoir une reproductibilité satisfaisante, est remplie avec 0.8 cm³ (épaisseur: 0.5 mm).

Quelques applications du dispositif

1. Étalonnage relatif de solutions radioactives

— Reproductibilité: pour systématiser les mesures on utilise toujours un volume de solution de 2 cm³, quantité dépassant largement celle nécessaire pour être en couche épaisse pour les émetteurs peu énergiques. Une erreur de 1/20 cm³ entraîne une variation sur la mesure de 0.3% pour les émetteurs peu énergiques, de 0.1% pour le P³². Pour une solution déterminée les écarts par rapport à la valeur moyenne observés avec une série d'échantillons de même volume n'excèdent pas 0.5% pour S³⁵ et 0.2% pour P³².

La présence de quantités même très notables d'entraîneur ou de corps en solution n'affecte d'aucune façon cette reproductibilité. Ainsi des mesures ont pu être faites sur l'eau de mer, qui contient environ 40 g/litre de sels minéraux en solution. Il est simplement nécessaire de faire un étalement préalable, qui ne présente aucune difficulté particulière. A partir de solutions de ce genre il n'eut pas été possible de préparer des sources solides d'activités reproductibles, la structure de telles sources, dans de telles conditions, n'étant pas homogène. La mesure de solutions organiques (p. ex. S³⁵ dans du toluène) ne présente également pas de difficulté.

— Efficacité du dispositif: on la définit comme étant le rapport: nombre d'impulsions enregistrées / nombre de rayons effectivement émis. Elle a été déterminée en partant de solutions de divers radioéléments préalablement étaillonnées au compteur 4 π (10⁻² μc/cm³). Les activités apparentes obtenues ont été exprimées en impulsions par minute.
On voit (figure 1 courbe 2) que la courbe de l'efficacité en fonction de l'énergie, relative au dispositif, se situe très nettement au-dessus de celle (courbe 5) qui correspond au compteur à liquide.

Figure 1

— Précision: l'erreur relative, résultant des erreurs sur la détermination du volume utilisé, sur l'étalonnage etc. varie de 3—6%/ suivant les cas. (Les activités spécifiques correspondantes sont $10^{-3}$ μс/см³ pour $^{14}C$ et $10^{-6}$ μс/см³ pour $^{32}P$.)

2. Détection de l'activité de solutions voisine de l'activité permise pour le radioélément considéré

Ce problème revêt de l'importance dans les contrôles d'eaux polluées par des substances radioactives. Il s'agit essentiellement de pouvoir contrôler par une méthode rapide, un grand nombre d'échantillons, et se rendre compte si l'activité est supérieure ou non à l'activité permise, pour le radioélément considéré.

Une méthode fréquemment utilisée consiste à préparer des sources après concentration préalable de la solution et à mesurer ensuite cette source à l'aide d'un compteur à fenêtre ordinaire mais à mouvement propre réduit par un dispositif à anticoïncidences. Cette façon de procéder soulève un certain nombre d'objections:
— efficacité faible (< 10%).
— la mesure n'est plus significative pour les basses énergies et lorsque la quantité de matière de la source est notable.
— méthode relativement peu rapide (2 à 3 heures par échantillon en raison des opérations de préparation de la source).

Le dispositif décrit nous semble échapper à ces objections. On peut en évaluer le seuil de détection. Si l'on désigne par:

m. p.: le mouvement propre du dispositif
A_g: l'activité apparente globale de l'échantillon
A_c: l'activité apparente globale, déduction faite du mouvement propre
σ étant l'écart quadratique moyen sur l'activité de la solution. Dans ces conditions \[ \sigma = \sqrt{2} \cdot \sigma^2 \] (m. p.) ou \( \sigma \) (m. p.) est l'écart quadratique sur le mouvement propre.

En faisant des déterminations de 30 minutes, on a
\[ \sigma = \sqrt{2} \times 30 \times \frac{11}{30} \approx 0.7 \text{ impulsions/minute}. \]
c'est-à-dire \( \sigma \approx 1 \) impulsions/minute.

L'activité décelable minimum est celle qui dans ces conditions donnera une impulsion/minute. Pour un mouvement propre de 3 impulsions/minute, le seuil de détection serait abaissé à 0,4 impulsions/minute.

A partir de cette convention et en utilisant la courbe de l'efficacité en fonction de l'énergie (figure 1) nous avons calculé les seuils de détection pour un certain nombre de radioéléments. Dans le tableau ci-dessous nous avons fait figurer outre les seuils de détection, les activités limites permisibles et pour chaque radioélément le rapport entre ces deux grandeurs. Les valeurs indiquées des activités permisibles sont celles qui ont récemment été recommandées par Morgan et adoptées par la Commission Internationale de Protection contre les Radiations Ionisantes (Juillet 1959).

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Seuil ( \sigma ) (µe/cm²)</th>
<th>Activité permissible A. P. (µe/cm²)</th>
<th>A. P./s</th>
</tr>
</thead>
<tbody>
<tr>
<td>C¹⁴</td>
<td>6.10⁻⁵</td>
<td>3.10⁻³</td>
<td>50</td>
</tr>
<tr>
<td>S²⁵</td>
<td>5.10⁻⁵</td>
<td>6.10⁻⁴</td>
<td>12</td>
</tr>
<tr>
<td>Ca⁴⁵</td>
<td>10⁻⁵</td>
<td>9.10⁻⁵</td>
<td>9</td>
</tr>
<tr>
<td>Co⁹⁶</td>
<td>6.10⁻⁶</td>
<td>3.10⁻⁴</td>
<td>50</td>
</tr>
<tr>
<td>I¹³¹</td>
<td>2.10⁻⁶</td>
<td>10⁻⁵</td>
<td>5</td>
</tr>
<tr>
<td>Co⁵⁷</td>
<td>2.10⁻⁶</td>
<td>2.10⁻⁴</td>
<td>100</td>
</tr>
<tr>
<td>Cs¹³⁷</td>
<td>8.10⁻⁷</td>
<td>10⁻⁴</td>
<td>125</td>
</tr>
<tr>
<td>P³²</td>
<td>5.10⁻⁷</td>
<td>2.10⁻⁴</td>
<td>400</td>
</tr>
<tr>
<td>Sr⁸⁰</td>
<td>6.10⁻⁷</td>
<td>10⁻⁶</td>
<td>1,7</td>
</tr>
</tbody>
</table>

On voit que le rapport A. P./s est toujours supérieur à 1, même pour Sr⁸⁰. Il est donc possible de mesurer directement, sans concentration préalable, au niveau de l'activité permissible tous les isotopes mentionnés ci-dessus. Cette
détermination, pour un temps global de manipulation de 35 minutes (préparation de la source: 5 min. — mesure: 30 min.), sera faite avec une précision de 10% environ.

3. Dosage d'une impureté radioactive au sein d'une solution d'un autre radioélément

Lorsqu'une solution d'un radioélément comprend de faibles quantités d'un autre radioélément, le repérage de cette impureté, l'évaluation de sa quantité sont des problèmes qui se posent quelquefois. Lorsque l'énergie β de l'impureté est nettement supérieure à celle des β du radioélément fondamental et lorsqu'elle efficacité du dispositif dépend de l'énergie du rayonnement, la présence de l'impureté peut fausser considérablement, la mesure de l'activité de la solution.

Pour aborder ce problème, une modification a été apportée au dispositif décrit plus haut. On utilise, placés dans la même enceinte, 3 compteurs: 2 à une fenêtre (C₁ et C₃) et un à deux fenêtres (C₂). La solution est placée dans la même cuvette que précédemment. L'ensemble du dispositif est représenté sur la figure 2.

Désignons par A₁, A₂, A₃ les activités apparentes globales (corrigées du mouvement propre) d'une solution donnée correspondant respectivement aux compteurs C₁, C₂, C₃.
Nous avons déterminé expérimentalement pour une série d'isotopes la valeur du rapport \( R = \frac{A_1}{A_2 + A_3} \). La valeur de ce rapport dépend naturellement des caractéristiques du dispositif (distances cuvette-compteurs, épaisseur des fenêtres etc...). Le résultat de ces déterminations est représenté sur la figure 3.

Connaissant la valeur de ce rapport pour le radioélément considéré il est possible de répéter la présence d'une faible quantité d'une impureté émettant un rayonnement plus énergique que celui du premier corps. La présence de cette impureté modifie en effet la valeur du rapport.

Ainsi l'addition à une solution de S\(^{35}\) d'une quantité de P\(^{32}\) dont l'activité totale est 0.1\% de celle du soufre, fait passer le rapport de 1.83\% à 2.54\% (variation de 39\%). Si l'on admet qu'une variation de 8\% de ce rapport est observable, la teneur décelable de l'impureté P\(^{32}\) dans une solution de S\(^{35}\) est de 2/10.000.

Ce procédé ne permet naturellement pas de séparer deux émetteurs d'énergies voisines. Mais dans ce cas, les efficacités correspondant aux deux rayonnements étant voisines, l'erreur provoquée par la présence d'une faible quantité d'impureté sera proportionnelle à cette quantité et non plus considérable comme dans le cas d'une impureté beaucoup plus énergique que le radioélément fondamental. Pratiquement, cette méthode ne convient qu'à la recherche d'impuretés radioactives au sein d'émetteurs \( \beta \) de basse énergie.
II. 2. 6. LOW-LEVEL β AND γ COUNTING IN THE REGION 0—10 DISINTEGRATIONS PER MINUTE

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Abstract

Measurements of levels of radioactivity in the region of 0—10 disintegrations per minute are becoming increasingly useful in the standardization of radionuclides as well as in other branches of science such as geology and medicine. For example, there is frequent need for low-level counting in the field of biological and tracer applications, particularly, in those cases in which the use of a higher level of radiation would alter the course of the mechanism or process under study.

Frequently such measurements must be made under conditions in which the sample possesses an activity of only 1% of the normal, unshielded background radiation. At other times, an additional requirement is imposed because the radionuclide may have a very short half-life.

There are three general ways in which the reliability of low-level measurements can be increased: more efficient counters, smaller background corrections and more stable, noise-free electronic systems.

In this paper, comparative tests are described of the relative merits of three different types of guard (anti-coincidence) counters in reducing the background. Measurements were made of: (a) umbrella, (b) annular, and (c) scintillating plastic igloo.

The increasing difficulty of obtaining activity-free materials for construction of counters is a matter of some concern. Data are presented for the range of activities of materials available commercially. Stockpiling of such materials, preferably of pre-War II origin is rapidly becoming a necessity.

The stability of the electronic system is a prime requisite for low level counting, particularly to avoid the counting of spurious pulses. A system is described in which a slow, measured rate of pulses is fed into the electronics, passed through the amplifier and deducted from the gross output of the register. In general, transistorized circuitry including pre-amplifiers and power supplies are more satisfactory than conventional vacuum tube devices.

Comptage β et γ d'échantillons de faible activité
(0 à 10 désintégrations par minute)

La mesure des niveaux de radioactivité de l'ordre de 0 à 10 désintégrations par minute devient de plus en plus utile dans la normalisation des radionuclides de même que dans d'autres domaines scientifiques, tels que la géologie et la médecine.

Ainsi le comptage de faible intensités est souvent nécessaire en biologie et dans l'emploi des indicateurs, surtout lorsque l'emploi d'un niveau de rayonnement plus élevé modifierait le déroulement du mécanisme ou du processus étudié.

Ces mesures portent souvent sur des échantillons ayant une activité de 1% seulement de l'activité de fond normale (sans écran). Dans d'autres cas, il faut tenir compte en outre du fait que le radionuclide a une période très courte.
Il y a trois manières générales d’augmenter la précision des mesures de faibles intensités: efficacité plus grand des compteurs, diminution des corrections nécessitée par l’activité de fond et emploi de circuits électroniques plus stables et plus silencieux.

Le mémoire décrit des essais comparés qui font ressortir les avantages relatifs que présentent trois types différents de compteurs d’anticoincidence pour la réduction de l’activité de fond: a) compteurs «champignons»; b) compteurs annulaires; c) dômes en matière plastique à scintillations. Il devient de plus en plus difficile d’obtenir des matières non actives pour la construction des compteurs. L’auteur donne des indications sur la gamme d’activité des matières que l’on peut se procurer dans le commerce. Le stockage de ces matières, qui doivent de préférence être d’avant-guerre, devient de plus en plus nécessaire. La stabilité du circuit électronique est indispensable au comptage de faible intensité, afin surtout d’éviter les impulsions parasites. Le mémoire décrit un dispositif dans lequel des impulsions lentes sont mesurées, puis introduites dans l’appareillage électronique; elles passent ensuite par l’amplificateur et sont enfin défalquées du chiffre brut indiqué par le numérateur. D’une manière générale, les circuits à transistors, y compris les préamplificateurs et alimentations, sont plus satisfaits que les dispositifs classiques fonctionnant avec des tubes à vide.

Измерение β- и γ-излучений малой мощности (0—10 распадов в минуту)

Измерение радиоактивности на уровне в области, содержащей от 0—10 распадов в минуту, становится все более полезным при стандартизации радиоактивных изотопов, а также в таких других отраслях науки, как геология и медицина например. Часто появляется необходимость в подсчете излучений малой мощности в области биологии и при исследовании методом меченых атомов, в частности, в тех случаях, когда использование радиации большой мощности может изменить ход работы аппарата или оказывать влияние на изучаемый процесс.

Часто такого рода измерения должны проводиться в условиях, когда образцы обладают активностью, превышающей обычный неэкранированный фон радиации лишь на 1%. В других случаях появляются дополнительные требования вследствие того, что радиоактивные изотопы могут иметь очень короткий период полураспада.

Имеются три основных пути увеличения надежности измерений радиации малой мощности: более совершенные счетчики, меньшие поправки на фон и более надежные свободные от помех электронные системы.

В данном документе описываются сравнительные эксперименты, связанные с изучением относительных качеств трех различных типов защищенных счетчиков (антисовпадений) для уменьшения фона. Измерения производились с помощью следующих типов счетчиков: a) топцевым, б) цилиндрическим, с) сцинтилляционным 4 л.

В документе уделяется некоторое внимание трудностям получения свободного от активности материала, используемого для производства счетчиков. В нем приводятся данные о материалах с различным содержанием активности, которые можно свободно приобрести. Накопление таких материалов, полученных преимущественно еще до второй мировой войны, быстро превращается в необходимость.
Надежность электронной системы является необходимой для измерений излучения малой мощности, в частности, для того, чтобы исключить подсчет побочных импульсов. В документе дается описание системы, в которой редкие импульсы, после измерения их скорости, направляются в электронную цепь, проходят через усилитель и выводятся через основной выходной канал счетного механизма. В основном транзисторная схема, включающая предварительное усиление и (электро)питание, является более удовлетворительной, чем схема, использующая вакуумную лампу.

Recuento β y γ de muestras de baja actividad
(0 a 10 desintegraciones por minuto)
La medición de niveles de radiactividad en el intervalo comprendido entre 0 y 10 desintegraciones por minuto presenta cada día mayor utilidad para la normalización de radionúclidos, así como en otras ramas de la ciencia tales como la geología y la medicina.
Por ejemplo, resulta a menudo necesario efectuar el recuento de las radiaciones emitidas por muestras de baja actividad en la esfera de las aplicaciones biológicas y de los trazadores, especialmente en aquellos casos en que el empleo de una radiactividad más elevada alteraría la marcha de la operación o del proceso que se estudia.

Con frecuencia, es preciso realizar esas mediciones en condiciones en que la actividad de la muestra apenas represente el 1% de la radiación de fondo en ausencia de toda protección. En otros casos, se impone otra limitación por el hecho de que el radionúclido tiene un período de semidesintegración muy corto.

En general, pueden seguirse tres procedimientos para aumentar la exactitud en el recuento de radiaciones de bajo nivel: utilización de contadores más eficientes, correcciones del fondo más pequeñas y empleo de sistemas electrónicos más estables y libres de interferencias.

En esta memoria se comparan los resultados de las pruebas llevadas a cabo con el propósito de averiguar las ventajas relativas de tres tipos distintos de contadores de anticoincidencia en lo que atañe a la reducción del fondo. A tal efecto, se efectuaron mediciones con contadores: a) tipo hongo, b) anular y c) "igloo" plástico de centelleo.

Causa alguna preocupación la creciente dificultad de obtener materiales inactivos para la construcción de contadores. Se ofrecen datos sobre los niveles de radiactividad de los distintos materiales que pueden adquirirse en el comercio. Se está agudizando rápidamente la necesidad de constituir reservas de dichos materiales, preferentemente de origen anterior a la segunda guerra mundial.

Para el recuento de las radiaciones procedentes de muestras de baja actividad es condición primordial la estabilidad del sistema electrónico utilizado, especialmente a fin de evitar el recuento de impulsos parásitos. Se describe un procedimiento que consiste en alimentar el sistema electrónico con impulsos generados a un ritmo determinado, que pasan por el amplificador y se descuentan del número bruto de impulsos registrado. En general, los circuitos equipados con transistores, inclusive en los preamplificadores y en las fuentes de tensión, son más satisfactorios que los dispositivos clásicos basados en el empleo de válvulas electrónicas.
I. Applications of Low-Level Counting

Measurements of levels of radioactivity in the region of 0 to 10 disintegrations per minute are becoming increasingly useful in the standardization of radionuclides as well as in certain other branches of science such as geology and medicine. There may be several reasons for making such measurements. For example, one may need to know the concentration of an impurity suspected of being present in a preparation being standardized, or one may be investigating a new species of radionuclide which can be produced only in very small quantities because of some nuclear limitation such as an extremely low capture cross-section for neutrons. Frequently such measurements must be made under conditions in which the sample possesses an activity of only 1% of the normal unshielded background.

The present paper does not cover the area of low-level counting in which the radioisotope is introduced as a gas into the counting volume, but deals with the more general case where the sample to be counted is in the solid form and may possess a wide range of $\beta$ or $\gamma$ energies.

II. Significance of the Figure of Merit

The figure of merit, $S^2/B$, is commonly used to compare various types of counters, where $S$ is the counting rate of the sample minus the background and $B$ is the background counting rate. It is misleading, however, to think of the figure of merit in terms of a single number, because of the complexity of the variables involved. Among the factors which affect the numerator, $S$, are the following:

- Activity of the preparation
- Solid angle subtended by the counter
- Window thickness
- Energy of the $\beta$ radiation
- Efficiency of counter detection for the type and energy of radiation entering the counter
- Sample thickness and backing material

Affecting the value of the denominator $B$ are:

- Thickness of shielding
  - primary
  - secondary
- Anti-coincidence measures
- Materials of construction
  - shielding
  - detector

It is easily possible to over-emphasize the value of reducing the background to improve the figure of merit, and although any attempt in this direction is worth while, it is by no means the only approach. Increasing the sample count rate by use of thinner windows, or by better geometry is even more valuable. The figure of merit of a counting arrangement may also be strongly dependent upon the energy of the $\beta$ radiation at low energies, and two counters which may be equally good for $\text{Sr}^{90}$ may differ appreciably in $S^2/B$ for $\text{C}^{14}$.

There is no "best" counter for low-level $\beta$ measurements. The maximum amount of time that can be devoted to counting the sample, together with the quantity of sample available, its specific activity and the number of samples
that can be counted at one time as well as the total number that must be counted all form part of the picture. Finally, two counting systems with an identical figure of merit may differ appreciably with regard to ease of sample preparation and insertion, facility of maintenance and repair, total weight, floor space and in initial cost. For these reasons, the values of \( S^2/B \) must be judged in conjunction with the factors described above when a laboratory is preparing to select low-level counting equipment.

Relative Figure of Merit of Two Counting Systems

If measurements of two counters are made under comparable conditions using the same radioisotope source, the relative figure of merit may be computed from the relationship:

\[
M = \frac{(S_1)^2/B_1}{(S_2)^2/B_2}
\]

While other relationships can be used, the above equation is generally preferred because of its simplicity.

III. Reduction of the Background Counting Rate

Much of the background counting rate can be eliminated by simple and multiple shielding, but the reduction of the small, residual rate is a very difficult problem.

For example, measurements were made in this laboratory by E. Balaam of the background counting rate of a DuMont 6292 photomultiplier tube and a Harshaw NaI(Tl) crystal measuring 1" \( \times \) 1—\( \frac{1}{2} " \). The data are presented in Table I.

It is difficult to reduce the 35 counts per minute remaining after 20 cm of iron bricks and 1.3 cm of mercury shielding are used. The effect of cosmic radiation (10 cpm) may be reduced further by anti-coincidence shielding; the effect of the \( ^{40} \text{K} \) contamination can be reduced by eliminating the mica support and the tube base; and the effect of \( ^{40} \text{K} \) in the NaI(Tl) crystal may be reduced by special purification of the reagents. The aluminum can and the \( \text{Al}_2\text{O}_3 \) were

<table>
<thead>
<tr>
<th>Shielding</th>
<th>Energy Range</th>
<th>Counts per Minute</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unshielded</td>
<td>Tube noise(^a) to infinity</td>
<td>1200</td>
</tr>
<tr>
<td>20 cm iron bricks</td>
<td>ditto</td>
<td>90</td>
</tr>
<tr>
<td>20 cm iron bricks plus 1.3 cm mercury</td>
<td>ditto</td>
<td>55</td>
</tr>
<tr>
<td>ditto</td>
<td>80 keV(^b) to infinity</td>
<td>45</td>
</tr>
<tr>
<td>ditto</td>
<td>80 keV(^b) to 2.5 MeV</td>
<td>35</td>
</tr>
</tbody>
</table>

\(^a\) Tube noise approximately 6 keV
\(^b\) Above Hg X-rays
Sources of the Remaining 35 Counts per Minute Background

<table>
<thead>
<tr>
<th>Source</th>
<th>Counts per minute</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cosmic radiation</td>
<td>10</td>
</tr>
<tr>
<td>Phototube contamination, K$_{40}$</td>
<td>7 to 10</td>
</tr>
<tr>
<td>K$_{40}$ in the NaI crystal (betas)</td>
<td>4</td>
</tr>
<tr>
<td>Aluminum crystal can, Al$_2$O$_3$ powder,</td>
<td>6</td>
</tr>
<tr>
<td>and glass face plate</td>
<td></td>
</tr>
<tr>
<td>Residual, unresolved activity</td>
<td>5 to 8</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td><strong>32 - 38</strong></td>
</tr>
</tbody>
</table>

found to contain 0.6 disintegrations per minute of radium per gram of Al content.

Some further reduction can be made if the glass face of the phototube is replaced by quartz, but it is equally important to note that the source of the quartz should also be examined.

Measurements made in this laboratory by R. H. Smith using neutron activation analysis showed a wide variation in the uranium content of two samples of quartz.

<table>
<thead>
<tr>
<th>Source of Quartz</th>
<th>Uranium Content, parts per million</th>
</tr>
</thead>
<tbody>
<tr>
<td>Manufacturer A</td>
<td>0.1</td>
</tr>
<tr>
<td>Manufacturer B</td>
<td>0.00006</td>
</tr>
</tbody>
</table>

If one assumes that in both cases the uranium in the quartz was of normal isotopic composition and that the quartz window in a phototube weighs approximately 100 grams, one would find for the quartz from Manufacturer A, a total of 15 α disintegrations per minute from the uranium plus chain daughters contributing to the γ activity. This is not a carte blanche approval of the product of Manufacturer B because his quartz may well contain other impurities equally undesirable.

IV. Materials of Construction

The problem of finding suitable materials of construction will get worse rather than better as time goes on, and serious thought has been given to the advisability of stockpiling uncontaminated materials. Many commercial manufacturers of radiation-detection instruments have been forced to go to considerable efforts to set up quality-control analyses for incoming materials. The problem is further aggravated by the need for producing reliable instruments which have lower and lower background counting rates for use in trace analyses such as nuclear fallout, activation analyses, etc.
Typical levels of contamination which have been measured are listed in Table III.

The radioactivity of stainless steel is an interesting case in point. Stainless steel produced prior to World War II shows a satisfactorily low activity, but that produced subsequent to approximately 1953 shows a relatively high activity because of the introduction of newer alloys containing rare-earth elements added to improve the properties of the stainless steel. The rare-earth elements contained a rather large amount of activity, usually thorium.

<table>
<thead>
<tr>
<th>TABLE III</th>
</tr>
</thead>
<tbody>
<tr>
<td>Typical Levels of Radioactivity of Materials of Construction</td>
</tr>
<tr>
<td>Total observed counts over background from 80 keV — 1 MeV on 3&quot; diam × 3&quot; crystal NaI (TI1)</td>
</tr>
</tbody>
</table>

| Material | 1.0 cpm/gram |
| Electrolytic copper foil | 0.02 cpm/gram |
| Copper, oxygen free | 35.4 cpm |
| 2" Photomultiplier tube socket | 0.08 cpm/gram |
| 60-40 Solder | 32.2 cpm |
| Glass photomultiplier tube base with leads RCA 6655 A | 4.2 cpm |
| RCA 6655 A | 72 cpm |
| Photomultiplier tube cage of RCA 6655 | <.3 cpm |
| Apiezon “Q” wax | 1.5 cpm/gram |
| Apiezon W 40 wax | <.1 cpm/gram |
| Bakelite | 0.08 cpm/gram |
| Lug posts | 3.5 cpm/100 |
| Scotch electrical tape | 0.05 per cm² |
| Scotch tape, regular | 0.2 per cm² |
| Modern lead brick | 50 — 250 |
| Modern lead brick, sandblasted | 10 — 20 |
| Steel brick | 50 — 500 |
| Steel brick, sandblasted | 5 — 25 |
| Mica, Indian Ruby 10 mg/cm² | 20 — 40 |

It is interesting to note that the body of some $\beta$ counters are being made of plastic to get away from the contamination; rather than metal. Obviously the plastic must be derived from “old” carbon such as petroleum or coal, for otherwise with “living” carbon the interior of the counter acts like an internal source of $\beta$ radiation which emits approximately 3 disintegrations per minute per 100 square centimeters of internal exposed area. Contemporaneous carbon possesses an activity of approximately 15 disintegrations per minute per gram.

V. Stockpiling of Uncontaminated Materials

The necessity of having to test each batch of construction materials is wasteful of manpower, time-consuming and quite costly; on occasion a year or more has been required to construct a duplicate of a satisfactory counting assembly unless a sufficient quantity of “activity-free” material had been previously stockpiled as the prototype was being assembled.
Many opinions have been expressed both pro and con as to the desirability of setting aside or stockpiling large quantities of activity free materials. Attention has been drawn to the relatively large quantities of pre-World War II steel, for example, scattered around the United States and presumably throughout the world in the form of old steamships, locomotives, railroad rails, etc. Indeed one admires covetously the "mothball" fleet of World War II ships with this thought in mind. Many of these ships contain lead ballast and other valuable materials such as copper, zinc, and aluminum.

The Subcommittee on Radiochemistry of the National Research Council (USA) with Dr. W. W. Meinke as its Chairman is investigating the desirability of stockpiling various materials, the quantities that might be needed, and the permissible levels of contamination. The results of a questionnaire dispatched to approximately 500 interested persons and organizations are being studied.

VI. Radiochemical Purity of Reagents

Closely allied to the problem of materials of construction is that of contaminated reagents. Several cases have been reported (2) in which radioactive contamination has caused serious difficulties in radiochemical analyses. Sodium hydroxide, for example, has been found to contain $10^{-11}$% of radium; ceric salts have been so contaminated with thorium as to interfere with their use as oxidimetric standards.

In this laboratory we have repeatedly encountered traces of uranium in various reagents. Not only does the concentration of uranium vary, but the isotopic ratio of $^{235}$U/$^{238}$U as well. These impurities are listed in Table IV in units popularly called a "smidgin" or $10^{-9}$ grams.

<table>
<thead>
<tr>
<th>Uranium content in smidgins/liter *</th>
</tr>
</thead>
<tbody>
<tr>
<td>H$_2$O distilled (average)</td>
</tr>
<tr>
<td>H$_2$O distilled twice from quartz</td>
</tr>
<tr>
<td>HNO$_3$ - reagent grade</td>
</tr>
<tr>
<td>Fuming HNO$_3$ - commercial</td>
</tr>
<tr>
<td>Fuming HNO$_3$ - redistilled</td>
</tr>
<tr>
<td>HF - commercial</td>
</tr>
<tr>
<td>HClO$_4$ - reagent</td>
</tr>
<tr>
<td>HClO$_4$ - distilled</td>
</tr>
</tbody>
</table>

* 1 smidgin = $10^{-9}$ gm

Cerium oxide has been found to contain $0.04\%$ Th$^{232}$; beryllium (3) and zirconium oxides also contain Th and U activities. The situation is becoming quite annoying for laboratories performing a combination of radiochemistry and low-level counting.

Some of these impurities, such as thorium in cerium are normally present; others such as U in HF and HNO$_3$ have obviously been introduced in recent times. In some cases, particularly in HF, the "reagent blank" for U may be several times that in the sample.
It is interesting at this point to note the enormous differences in the relative sensitivities between radiochemical and conventional chemical techniques. What would be accepted as "chemically pure" or "reagent grade" materials in conventional chemistry may be totally inadequate for radiochemical work, particularly in activation analyses.

The situation is not unlike that existing around the turn of the century, when chemical reagents contained many impurities. Older chemists will remember that one chemical company took the initiative in purifying and marketing "special" reagents and the word "Kahlbaum" became synonymous with chemical purity.

It is obviously a waste of scientific manpower to require each radiochemist to purify his own reagents and it is virtually impossible for him to be assured of their purity. The Subcommittee on Radiochemistry of the National Research Council and the National Bureau of Standards are actively working toward the radiochemical purification of reagents. There is a very real danger that unless such specially purified reagents are made available, many of the low-level radiochemical analyses of foods, drinking waters and environmental contamination will be misleading, if not worthless.

VII. Comparison of β Counting Systems

The figures of merit of six β end-window counting systems together with the recommended shielding were measured by R. D. Davis of this Laboratory using four solid radioisotope sources mounted on planchets. The β energies of the isotopes used ranged from 0.158 MeV for C\textsuperscript{14} to 2.27 MeV, for Sr\textsuperscript{90}-Y\textsuperscript{90}.

Photographs of the counters and counting arrangements used are shown in figures 1, 2, and 3. The counters tested were as follows:

<table>
<thead>
<tr>
<th>Counter No.</th>
<th>Type</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Aluminum end-window, 1.6 mg/cm(^2); flow counter, proportional region; methane gas.</td>
</tr>
<tr>
<td>2</td>
<td>Body of cast Epon resin; Mylar end-window 0.9 mg/cm(^2); Geiger region; filling: helium plus organic quench.</td>
</tr>
<tr>
<td>3</td>
<td>Mica end-window, 1.5 mg/cm(^2); Geiger region; filling: neon, argon plus halogen quench; placed inside hollow anode guard counter.</td>
</tr>
<tr>
<td>4</td>
<td>Same as counter 3, except for 10.0 mg/cm(^2) stainless steel window.</td>
</tr>
<tr>
<td>5</td>
<td>Aluminized Mylar end-window 0.9 mg/cm(^2); Geiger region; filling: helium plus organic quench; flow counter.</td>
</tr>
<tr>
<td>6</td>
<td>Mica end-window 2.5 mg/cm(^2); Geiger region; filling: helium plus organic quench; similar to counter No. 5, except for window thickness.</td>
</tr>
</tbody>
</table>

The physical dimensions and other data such as the length of the plateau are listed in Table V. The background counting rates were determined under various conditions of shielding, and with the proper amount recommended for use with each particular counter. These data are given in Table VI. Table VII lists the radioisotopes used in this study.
Figure 1
Counter No. 1. Methane end-window flow counter; 1.6 mg/cm², Al window, CH₄ filling.

TABLE V
Description of β End-Window Counters Tested

<table>
<thead>
<tr>
<th>Counter No.</th>
<th>Window mg/cm²</th>
<th>Filling Gas</th>
<th>Eff. Diam. of Cathode</th>
<th>Operating Voltage</th>
<th>Plateau</th>
<th>Slope %/100V</th>
<th>Resolving Time μsec</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.6 Al</td>
<td>CH₄</td>
<td>3.3 cm</td>
<td>4300</td>
<td>750</td>
<td>0.5</td>
<td>3</td>
</tr>
<tr>
<td>2</td>
<td>0.9 Mylar Al Coated</td>
<td>He + organic quench</td>
<td>3.3 cm</td>
<td>1400</td>
<td>250</td>
<td>0.5</td>
<td>150</td>
</tr>
<tr>
<td>3</td>
<td>1.5 Mica</td>
<td>Ne, A, + halogen quench</td>
<td>2.0 cm</td>
<td>550</td>
<td>550</td>
<td>3</td>
<td>150</td>
</tr>
<tr>
<td>4</td>
<td>10.0 Stainless Steel</td>
<td>do</td>
<td>2.8 cm</td>
<td>550</td>
<td>550</td>
<td>3</td>
<td>200</td>
</tr>
<tr>
<td>5</td>
<td>0.9 Mylar Al</td>
<td>He + organic quench</td>
<td>3.3 cm</td>
<td>1250</td>
<td>150</td>
<td>2</td>
<td>100</td>
</tr>
<tr>
<td>6</td>
<td>2.5 mica</td>
<td>He + organic quench</td>
<td>3.7 cm</td>
<td>1250</td>
<td>150</td>
<td>2</td>
<td>100</td>
</tr>
</tbody>
</table>

* See Figures 1, 2, and 3.
Counter No. 2. Two cast Epon counters, 0.9 mg/cm², aluminized Mylar window; He + organic quench filling; the Geiger coincidence umbrella is shown above.

TABLE VI

Observed Background Counting Rates in Counts per Minute

<table>
<thead>
<tr>
<th>No. Counter</th>
<th>Shielding Used</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>3.5 cm Pb</td>
</tr>
<tr>
<td>1</td>
<td>14.5 cpm</td>
</tr>
<tr>
<td>2</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td></td>
</tr>
</tbody>
</table>
Left to right: (a) Counter No. 2. Cast Epon counter; 0.9 mg/cm², aluminized Mylar window, He + organic quench filling; (b) Counter No. 3 (1.5 mg/cm² window) and Counter No. 4 (10.0 mg/cm² window). Spherical anode beta counter, mica window, Ne + A + halogen quench with hollow anode guard counter shown above; and (c) Counter No. 5. Flow counter 0.9 mg/cm², aluminized Mylar window, He + organic quench filling. Counter No. 6 (not shown) is similar to Counter No. 5 except for a window thickness of 2.5 mg/cm².

**TABLE VII**

**Isotope Sources Used**

<table>
<thead>
<tr>
<th>Source</th>
<th>Beta Energy MeV</th>
<th>Details of Preparation</th>
</tr>
</thead>
<tbody>
<tr>
<td>C¹⁴</td>
<td>0.158</td>
<td>BaC¹⁴O₃; 30.18 mg/cm² layer deposited on filter paper with Al backing; covered with Mylar film approximately 0.9 mg/cm² thick.</td>
</tr>
<tr>
<td>Tc⁹⁹</td>
<td>0.29</td>
<td>Plated on Cu with brass backing; sprayed with Krylon, covered with Mylar approximately 0.6 mg/cm² thick.</td>
</tr>
<tr>
<td>Cl⁹⁶</td>
<td>0.71</td>
<td>Stippled AgCl source on Cu disc with Al backing; aluminized Mylar cover, approximately 0.9 mg/cm² thick.</td>
</tr>
<tr>
<td>Sr⁹⁰-Y⁹⁰</td>
<td>0.54, 2.27</td>
<td>SrCl₂ stippled on Cu disc with brass backing; pliofilm cover approximately 0.6 mg/cm² thick.</td>
</tr>
</tbody>
</table>
The comparative figures of merit obtained for each counter relative to Counter No. 1, arbitrarily chosen as a reference are shown in figure 4.

VIII. Low-Level $\gamma$ Counting

The general techniques of low-level $\gamma$ detection are similar to those of $\beta$ detection only insofar as in both cases the elimination of activity from the counter material and the reduction by shielding of the ambient $\gamma$ flux are of prime importance. Considerable advantage can be obtained in $\beta$ counter construction by the reduction of the counter sensitivity to $\gamma$-rays. For the $\gamma$ counter this technique is not applicable, but the discrete energy of the $\gamma$ and the

Figure 4

Relative figure of merit for various low-level, $\beta$ end-window counting systems.
availability of detectors of high resolution with respect to γ energy make possible the utilization of spectrometric analysis. For low-level spectrometry, where we are concerned with the photopeak portion of the γ spectrum, it is obvious that statistical separation of the sample counts from the background is dependent on the resolution of the spectrometer system and if, in addition, the instrument is to be used to separate the radiations from complex samples, the resolution assumes added importance. The expression for the figure of merit applicable to γ spectroscopy is:

\[ M = \frac{k \epsilon R^2}{B} \quad \text{where } k \text{ is a proportionality factor} \]

\( \epsilon \) is the counter efficiency
\( R \) is the resolution; defined here as inversely proportional to the full width of the photopeak at 1/e height
\( B \) is the background.

The background is essentially proportional to the crystal volume, and it can be reduced by reducing the crystal size. The efficiency on the other hand, increases with the crystal volume. Crystal resolutions, as defined above, decrease with the size of the crystal. The situation with regards to the resolution is complicated by the fact that, in addition to the inherent statistical resolution of the crystal due to conversion efficiency and light attenuation, for crystals demanding large area phototubes, the resolution of the phototube, due to cathode non-uniformities, becomes comparable with that of the crystal. Typical figures for resolutions (for 660 keV Cs\textsuperscript{137} line) of crystal-phototube assemblies are:

- 3" X 3" diameter to 3" phototube = 6%
- 5" X 5" diameter to 5" phototube = 10%

The resolution of the spectrometer is not entirely governed by the resolution of the detector assembly. If the counting times exceed a few hours, the drifts encountered in the amplifiers, analyzer and especially in the high-voltage supply to the phototube appreciably increase the spread of the photopeak. One method of controlling these drifts which avoids the extreme of precise stabilization of all of the components is described later.

In the design of a low-level γ spectrometer by R. M. Main and Gordon Oescher of this laboratory it became necessary to maximize the so-called figure of merit of the system. The conditions are such that varying the crystal dimension to increase the efficiency adversely affects both the resolution and the background. When the figure of merit is optimized with respect to the crystal dimensions for various energies, the crystal size must vary radically over the normally important region 60 keV to 2.0 MeV. No single crystal is reasonably applicable over the entire region. In this laboratory the crystal sizes which have been chosen are:

- 60—300 keV 2" diameter × ½"
- 250—750 keV 2" diameter × 1—½"
- 600—2500 keV 3" diameter × 3"

In the construction of these detectors extreme care has been exercised in the choice of materials. Special crystals with a low potassium content (approximately 5 ppm) were obtained from Harshaw Chemical Company, coupled to quartz-faced phototubes with the bases removed.
All of the materials to be used within the shielded cavity were first counted on a low-level $\beta$ counter. After the $\gamma$ detector was constructed, using materials which were as low as possible in $\beta$ activity, samples of the materials were $\gamma$ counted. Those which produced detectable counts were eliminated from the system. The exception was aluminum oxide, known to be slightly active, which was used as the crystal reflector. In this case background was sacrificed slightly in favor of the increased resolution possible using this material in comparison with all other materials tested.

An example of the counting conditions obtainable with the 3" diameter $\times$ 3" crystal are as follows:

<table>
<thead>
<tr>
<th>Energy</th>
<th>Bkgd/10 keV Width</th>
<th>Photopeak Efficiency</th>
<th>Resolution</th>
<th>Bkgd under Photopeak, cpm</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5 MeV</td>
<td>0.8</td>
<td>15.5</td>
<td>6.5% 32 keV</td>
<td>2.5</td>
</tr>
<tr>
<td>1.0</td>
<td>0.4</td>
<td>6.5</td>
<td>5.0% 50 keV</td>
<td>2.0</td>
</tr>
<tr>
<td>1.5</td>
<td>0.25</td>
<td>2.5</td>
<td>4.5% 67 keV</td>
<td>1.7</td>
</tr>
</tbody>
</table>

It should be emphasized that the activity found for each of the materials of construction applies only to that piece of material and that every new material, no matter what its source, must be suspected until measured.

IX. Reduction of Spurious Counting

As mentioned previously, when both the level of the sample and of the background are decreased, it becomes quite important that the detection system and the associated amplifier and circuitry remain electrically stable and that no spurious counts are introduced. This is particularly true if a complex $\gamma$ spectrum is being measured with a pulse-height analyzer using a photomultiplier tube as a detector, for in this case variations in the high-voltage supply, for example, can cause differences in the apparent pulse distribution and spread the counts over neighboring channels. In the previous section reference was made to the expression $M = kE^2R^2/B$, where $R$ is the resolution of the analyzer. Obviously improvements in the value of $R$ increase the usefulness of the instrument.

Such improvement in the resolution of the system as a whole can be achieved by better sensing elements, high-voltage stabilization, selection of low-drift components and use of feed-back and stabilization loops.

High-voltage stabilization is an important factor in improving the resolution of the system because the gain of the photomultiplier tube is controlled by the high voltage applied to the anodes of the tube.

Conventional methods of voltage stabilization involve use of constant-voltage transformers, voltage-regulator tubes and electronically regulated power supplies.

Other methods of stabilizing the high voltage involve the use of different feed-back loops. For example one can inject into the electronic system a simulated source of radiation whose energy (pulse-height) and intensity (number
of pulses per unit time) are known. This calibrating signal can be so adjusted as to periodically re-balance the high voltage.

In selecting equipment for a low-level counting laboratory, one must always bear in mind the use to which the instrument will be put.

X. Need for Low-Level Standards of Activity

It would be quite helpful to have available certified samples of low-level β and γ activities in the region of 0 to 10 disintegrations per minute for the calibration of low-level counting equipment. These low-level standards would be used in conjunction with the corresponding high-level standards of the same isotope. The latter are useful in checking the day-to-day performance of the counting system to detect quickly any major changes in performance. The low-level standards would be used, with sufficiently long counting times, to detect any spurious counting rate or other malfunction. It has been our experience that an instrument may appear to function well at 1000 cpm, for example, but that reliable measurements cannot be made in the region of 0 to 10 cpm even though the nominal background was both low and statistically constant.

XI. Trends in Low-Level Counting Instrumentation

In addition to the need for the stockpiling of materials of construction and for the radiochemical purification of reagents, various trends are apparent in the instrumentation used for low-level counting. Mention has been made of the stabilization of phototubes and amplifiers.

Transistorized circuitry is competing actively with designs using vacuum tubes. The higher costs of transistors, particularly of the specialized types needed, are in part off-set by their longer life and simpler and fewer components. Although transistors are themselves also temperature-sensitive, a transistorized instrument will operate at a lower temperature than the corresponding vacuum tube counterpart, and the lower heat output will not cause neighboring resistors and capacitors to drift as much in value.

Tunnel diodes and Zener diodes are becoming used as components of a few research type instruments.

Recent advances in solid-state detectors (4) such as silicon-phosphorus (5) and others (6) are opening new pathways toward greater resolution in pulse-height analysis and may well lead to families of very useful sensing elements.

REFERENCES

(5) HENSON, W., University of California Radiation Lab., private communication.
The authors describe a method of analysing the sequence overtime of cathode photoelectrons emitted during the scintillation induced by very low-energy particles. It is possible to distinguish the scintillation pulses involving two or more associated photoelectrons from the individual electrons which form the cathode dark current. With high-efficiency scintillators (e.g. ZnS*), the particle energy required to produce a pair of photoelectrons is less than 1 keV; particles of this energy are very efficiently distinguished. This method, whereby cooling the photomultiplier tube can be avoided, is suggested for tritium counting.

Méthode de comptage par scintillations de particules de très faible énergie (inférieure à 1 keV)

Les auteurs décrivent une méthode d'analyse de la succession dans le temps des photoélectrons cathodiques émis pendant la scintillation provoquée par des particules de très faibles énergies. Il est effectivement possible de distinguer les impulsions de scintillation comportant deux ou plusieurs photoélectrons groupés, des électrons individuels formant le "courant d'obscurité" (dark current) cathodique. Avec des scintillateurs de grande efficacité (par exemple ZnS*) l'énergie de la particule, nécessaire pour produire une paire de photoélectrons, est inférieure à 1 keV; des particules de cette énergie sont discriminées très efficacement. Cette méthode, qui permet d'éviter le refroidissement du photomultiplicateur, est proposée pour le comptage du tritium.

Метод сцинтилляционного счета частиц с низкой энергией (ниже 1 keV)

Авторы описывают один из методов анализа последовательности излучения по времени катодных фотоэлектронов при сцинтилляции, вызываемой частицами очень низких энергий. Вполне возможно отличить импульсы сцинтилляции, включающей два или несколько одновременно излучаемых фотоэлектронов, от отдельных электронов, образующих катодный темновой ток. На мощных сцинтилляторах (например, ZnS*) энергия частицы, необходимая для получения пары фотоэлектронов, ниже 1 keV; частицы этой энергии избираются весьма эффективно. Этот метод, позволяющий избегать охлаждения электронного фотоумножителя, предлагается для подсчета трития.

Recuento por centelleo de partículas de muy baja energía (inferior a 1 keV)

Los autores describen un método para analizar la sucesión en el tiempo de los fotoelectrones catódicos emitidos durante el recuento por centelleo de partículas de muy baja energía. De hecho, es posible establecer una distinción entre los impulsos de centelleo en los que intervienen dos o varios fotoelectrones agrupados y los debidos a electrones aislados que forman la "corriente oscura".
Scintillation counting of very low energy particles, such as $\text{H}^3\beta$, is not easily accomplished by discrimination of pulse amplitude, due to the high number of photomultiplier dark pulses of large amplitude spread.

Scintillation pulses, even made of several photoelectrons, are masked in the multitude of dark pulses having amplitudes in the same range.

This limitation can be avoided when time correlation between single photoelectron pulses belonging to the same scintillation is taken into account to discriminate against background made of pulses randomly spaced in time.

The classical method of doing this is to look a liquid scintillator, in which the radioemitter is dissolved, by a pair of photomultipliers and to count coincident pulses that arise when a single scintillation produces at least one photoelectron from each photocathode.

Uncorrelated dark pulses produce only a small number of random coincidences. It is usual to cool the system in order to make this number negligible.

The detection threshold of this method is, at least, 4 keV, that is two times the average energy to produce one photoelectron with a liquid scintillator.

In a new method we are developing, consideration of the time distribution law of primary electrons emitted in a single photomultiplier allows to measure the output pulses directly in number of primary electrons.

Discrimination of scintillation pulses against dark pulses is thus possible independently of the multiplication spread.

Practically, the scintillations are identified as those made, mostly, of two or more grouped primary electrons in a time of the order of the scintillation lifetime, whereas the background pulses are mostly made of single primary electrons randomly spaced in time.

For this method a kind of scintillator is needed with a long lifetime compared with the electronic resolving time.

Activated zinc sulphide is a particularly suitable material having a lifetime from few to several microseconds, following the type.

Its scintillation efficiency is such that affords, for instance, one photoelectron per $300 \sim 400$ eV, so that a detection threshold below 1 KeV can be reached with this method.

The experimental devices are presented in figure 1.

Output pulses from a photomultiplier are fed through a preamplifier to a circuit, we shall refer to as "multiplicity discriminator", that gives information about how many distinct pulses are coincident within time intervals of a pre-fixed duration.

Corresponding to each event of the desired kind, a counting pulse is generated and registered by a scaler.
An anti-coincidence gate, triggered by the preamplifier output, permits to discriminate against the counts originated by cosmic rays and external $\gamma$ radiation, as it will be described later.

The circuit diagram of the multiplicity discriminator is shown in figure 2. Section (A) is a pulse shaper that resolves and converts the photomultiplier output into standard pulses, each corresponding to a single primary electron.

The standard pulses are integrated in section (B) of the circuit, giving a resultant pulse of stepped shape, each of the subsequent steps corresponding to the arrival of a single electron pulse.

The operation is stopped after a time $\tau$ determined by a univibrator. Typical stepped shapes are shown in figure 3.

It is possible to preset the bias of a voltage discriminator, included in section (B), above the level of the first step in order to discard all single pulses and to count double or higher order pulses. By setting the bias above second step level, both single and double pulses are discarded and triple counted and so on.

According to the order of the preset level, the counting rate of random coincidences can progressively be reduced.

In our measurement the probability $\eta^3 \tau^2$ of triple random coincidences was negligible.

The circuit of section (C) provides a paralysis time, up to 500 $\mu$s, after each registered count, to avoid more counts being originated by the same scintillation.

The kind of photomultiplier we chose for the measurement is the E. M. I. 6255 S, with 13 multiplication stages.

The "S" type photocathode is an antimony-caesium cathode processed for low red sensitivity, having no exceedingly high thermoelectronic emission even at room temperature.

The photomultiplier circuit is shown in figure 4.
Figure 2
Circuit diagram of the multiplicity discriminator.
Typical response shapes of the multiplicity discriminator for a double and a triple event.

Figure 4
Photomultiplier circuit.
The effect of the graphite coating of glass envelope connected with the ground is both to absorb light and to catch positive ions generated in the last high-current stages before they reach the region of the cathode. Without this arrangement, a much larger number of multiple pulses probably due to delayed feedback to the cathode, were observed.

In order to choose the best operating conditions for our measurements, the amplitude spectrum of single pulses from the photomultiplier was studied.

In figure 5, the lower curve represents the integral counting rate versus discriminator bias with the photomultiplier in the dark. The upper curve is the counting rate with the photocathode exposed to a slight luminous flux from a tungsten lamp.

In both spectra the amplitudes of pulses due to primary electrons either from the cathode or from the first dynodes are mixed together.

The third plot is the difference between the counting rates measured in the previous conditions, and represents the contribution of primary photoelectrons only.

It is evident that above a certain bias practically all the photoelectrons are taken into account, whereas exceedingly high counting rates corresponding to lower bias are avoided.

All pulses above such a bias were accepted and analysed by our multiplicity discriminator.

Calibration of the bias scale in units of average cathode electron height has also been performed from measurements with a constant and known photoelectric current at the input of the multiplier.

![Figure 5](image)

Integral distribution of single electron pulses.
It is thus possible to illustrate the efficiency of the outlined method with a direct comparison between background discrimination versus the output pulse-height and versus the multiplicity order (i.e. the "input" pulse-height: see Table I).

<table>
<thead>
<tr>
<th>Amplitude discriminator level (measuring unit is the average output of a single cathode electron)</th>
<th>Multiplicity discriminator level (min. number of primary electrons per counted pulse)</th>
<th>Background counts cpm</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>( \tau = 20 \mu s )</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Amplitude discriminator</td>
</tr>
<tr>
<td></td>
<td></td>
<td>calculated</td>
</tr>
<tr>
<td>0.1</td>
<td>1</td>
<td>13000</td>
</tr>
<tr>
<td>1</td>
<td>2</td>
<td>3700</td>
</tr>
<tr>
<td>2</td>
<td>3</td>
<td>950</td>
</tr>
<tr>
<td>3</td>
<td>4</td>
<td>230</td>
</tr>
<tr>
<td>4</td>
<td>5</td>
<td>100</td>
</tr>
<tr>
<td>5</td>
<td>6</td>
<td>48</td>
</tr>
</tbody>
</table>

Scintillation measurements were made first with tritiated water samples. For this purpose the photomultiplier was supplied with a scintillation cell shown in figure 6.

A thin ZnS* layer was applied over the bottom of the cell, made of a plexiglass disc in optical contact with the photocathode.

The water sample was dropped on the ZnS* layer through a hypodermic needle and evacuated by syphon system.

The disc was covered with a nylon mesh to get the scintillator wetted and washed uniformly. With this system, once the scintillator was wet, the detection efficiency was found very constant and reproducible in the following measurements, independently of the flow rate.

In order to discriminate against cosmic radiation, the cover of the cell was internally coated with a plastic scintillator paint.

Due to the fast scintillation response of this phosphor with respect to the electronic resolving time, a crossing particle produced an output pulse, higher than those generated during the ZnS* scintillation, that was useful to trigger the anti-coincidence gate. In many cases this anti-coincidence method is effectual also against \( \gamma \) radiation, when the secondary electron is scattered from the ZnS* into the plastic and vice versa.

With the addition of a 5 cm lead shield the background of external radiation was reduced to a negligible level.

The background counting rates in various conditions are given in Table II a.

It can be noted that the counting rate of triple order pulses was small, the standard value being 6 cpm, and very little affected even by the excessive emission of single electrons (up to ~ 20,000 cpm) consequent to the exposure of ZnS* to daylight.

The detection efficiency for THO (~ 1 \( \mu \text{c/cc} \)) samples has been measured by working either with double level or with triple level counts (Table II b).
In the former case the detection efficiency was about twice, but a better useful-to-background ratio was possible with triple level counts.

Active water can be completely removed from the scintillator surface by flowing a few cc of clean water.

Replacement of samples is simple and quick, and does not affect the background counting rate.

We suggest that a method like that we used could be convenient for measuring the activity of liquid, or perhaps gaseous, samples in comparison with standard samples.

The advantage with respect to usual internal counting methods is to avoid the preparation of the scintillating solution and, therefore, to work with a scintillation efficiency independent of quenching agents. The trouble of C\textsuperscript{14} activity is also avoided. Photomultiplier cooling was not necessary.

Continuous measurement of liquid and gaseous flows is obviously obtainable with our device.

More generally, this kind of scintillation detector can be considered as a possible widowless particle counter useful at low-energy levels.
We have recently tried a kind of internal counting with intimate mixtures of THO, ZnS* and starch gel sandwiched between two plexiglass discs.

In preliminary measurements, the detection efficiency corresponded to a scintillation counting rate of the order of the background per $10^{-9}$ curies.

We hope to exceed this limit after various improvements in the preparation of samples.

A future extension of this technique, involving improvements in the electronic resolving time, may enable us to use inorganic crystals having a shorter scintillation lifetime [NaI(Tl), etc.] for the detection of very low-energy X-quanta.
II. 2. 8. THE COMPARATIVE ACCURACY OF THE $4\pi$ LIQUID SCINTILLATION COUNTING METHOD OF RADIOISOTOPE STANDARDIZATION

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Abstract

The accuracy of the $4\pi$ liquid scintillation counting method of standardizing $\beta$ emitters was compared to $4\pi\beta-\gamma$ coincidence counting for the nuclides Co$^{60}$, I$^{131}$ and Au$^{198}$. For P$^{32}$ the liquid counting results were compared to $4\pi$ proportional counting. The efficiency of the liquid scintillation counting method was found to be energy dependent, dropping to about 97.5% for Co$^{60}$ which was the lowest energy $\beta$ emitter investigated.

L’exactitude comparée de la méthode de comptage $4\pi$ à scintillateurs liquides pour l’étalonnage des radioisotopes

La précision de la méthode de comptage $4\pi$ à scintillateurs liquides pour l’étalonnage des émetteurs $\beta$ a été comparée au comptage par coïncidences $4\pi\beta-\gamma$ pour le Co$^{60}$, le I$^{131}$ et le Au$^{198}$. Dans le cas du P$^{32}$, les résultats du comptage au liquide ont été comparés à ceux du comptage $4\pi$ proportionnel. On a constaté que le rendement de la méthode de comptage à scintillateurs liquides variait en fonction de l’énergie émise et qu’il descendait à environ 97.5% pour le Co$^{60}$ qui, de tous les émetteurs $\beta$ étudiés, émet l’énergie la plus faible.

Сравнительная точность $4\pi$ жидкого сцинтилляционного метода подсчета стандартизируемых радиоизотопов

Точность $4\pi$ жидкого сцинтилляционного метода подсчета стандартизируемого $\beta$-излучателя сравнивалась с $4\pi\beta-\gamma$ методом подсчета на совпадениях для изотопов Co$^{60}$, I$^{131}$ и Au$^{198}$. Для P$^{32}$ результаты жидкого подсчета сравнивались с $4\pi$ пропорциональным методом подсчета. Эффективность метода жидкого сцинтилляционного метода подсчета оказалась зависимой от энергии и понизилась до 97.5% для Co$^{60}$, у которого найдена наименьшая энергия из исследованных $\beta$-излучателей.

Exactitud del método de recuento con centelleador líquido $4\pi$ para normalizar radioisótopos, comparada con la de otros métodos

El autor compara la precisión del método de recuento con centelleador líquido $4\pi$ para normalizar emisores $\beta$ con la del método de coincidencias $\beta-\gamma$ $4\pi$, para los siguientes núcleidos: Co$^{60}$, I$^{131}$ y Au$^{198}$. En el caso del P$^{32}$, confronta los resultados del primer método con los obtenidos mediante el recuento proporcional $4\pi$. Comprueba que la eficacia del método de recuento con centelleador líquido depende de la energía y desciende al 97.5%, aproximadamente, para el Co$^{60}$, que fue el emisor $\beta$ más débil que se investigó.
Introduction

The small-volume, liquid scintillation counter was first used by BELCHER\(^{(1)}\) for the determination of absolute disintegration rates. He obtained \(4\pi\) geometry by immersing dry source deposits sealed between thin films in the liquid scintillator. The problems of foil-absorption and self-absorption limit the accuracy of this method in exactly the same way as in the standard methods of \(4\pi\) counting using Geiger-Müller or proportional gas counters. The author\(^{(2)}\) showed how the problems of absorption could be eliminated by dissolving the radioactive substance in the liquid scintillator. Elaborate source preparation was unnecessary, and considerable amounts of carrier material could be tolerated in the radioactive solutions. However, it was still necessary to extrapolate a curve of counting rate against discrimination level to zero discrimination level to obtain the true disintegration rate. For all those radionuclides that have been investigated, counting conditions could be found for which the portion of the curve to be extrapolated was a straight line, rendering the extrapolation very precise. Consequently, good reproducibility was obtained in practice.

The validity of the extrapolation as giving the true disintegration rate has since been investigated for the nuclides Co\(^{60}\), I\(^{131}\), Au\(^{198}\) and P\(^{32}\). Except...
for P⁳², the source-disintegration rates found by extrapolation were compared to 4πβ-γ coincidence measurements (3) on these same sources.

The 4πβ-γ coincidence method (3) (4) (5) has many advantages over conventional β-γ coincidence counting. Amongst others, a very favourable ratio of true coincidence counting rate to accidental coincidence counting rate prevails, which makes it possible to use relatively long coincidence resolving times, hence eliminating any possibility of coincidence loss due to unequal delays in the channels. The method can also be applied with confidence to the standardization of many radionuclides having complex breakdown schemes.

In the case of P⁳², the specific activities of a number of solutions were determined by the liquid scintillation extrapolation method and then compared to the results of 4πβ proportional counting on the same solutions.

**Apparatus**

Counting cells containing about 12 ml of liquid scintillator could be placed directly on top of a vertically mounted E. M. I. 6097 S multiplier phototube. This liquid scintillation counter was part of an arrangement for 4πβ-γ coincidence counting, the γ counter being a NaI-scintillation spectrometer. Figure 1 shows the two scintillation counters adjacent to each other with part of the cast iron shield removed. The counters were set up in a refrigerator where the temperature was kept at about -15° C by thermostatic control.

Impulses from the counters were fed via cathode-follower preamplifiers to linear amplifiers followed by pulse-height selectors. The outputs from the pulse-
height selectors were fed to a coincidence unit constructed according to a circuit by SMALLER and AVERY (6). A coincidence resolving time of 4 $\mu$s was used throughout the work reported here. Care was taken to count only those impulses in the single channels which were on their way to the coincidence mixer, having overcome the bias in the pulse-shaping stages of the coincidence unit.

The amplifier used with the liquid scintillation counter was based on the design of CHASE and HIGINBOTHAM (7) and had very non-overloading characteristics. The discriminator following this amplifier was based on the design of FAIRSTEIN and PORTER (8).

For the extrapolation to zero impulse height, it was of the utmost importance to know the position of the zero point of the discriminator precisely. The discriminator used was checked by means of an impulse generator and a Tektronix 541 oscilloscope. As a further check the photoelectric peaks of a number of well-known $\gamma$-ray lines were measured using the amplifier and discriminator in conjunction with a good NaI scintillation counter. Figure 2 shows the linearity of the combination. The uncertainty in the discriminator zero point was not more than $\pm 1$ Volt.

The $4\pi$ proportional counter used for the absolute measurement of the $P^{32}$ solutions, was a flow counter of conventional design, having a “pill-box” shape, with straight anodes. The electronic auxiliary apparatus was the same as was used with the liquid scintillation counter.

**Method**

Samples of the aqueous radioactive solutions weighing about 0.04 g were dissolved in the liquid scintillator containing the appropriate carrier material for each radionuclide. Relevant data on the liquid scintillator are given in Table I. The aqueous sample dissolved rapidly in this scintillator after stirring for a few seconds with a thin glass rod.

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Chemical form</th>
<th>Concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>$C^{60}$</td>
<td>CoCl$_2$</td>
<td>50 mg/liter</td>
</tr>
<tr>
<td>$I^{131}$</td>
<td>NaI</td>
<td>10 mg/liter</td>
</tr>
<tr>
<td>$Au^{198}$</td>
<td>AuCl$_3$</td>
<td>50 mg/liter</td>
</tr>
<tr>
<td>$P^{32}$</td>
<td>H$_3$PO$_4$</td>
<td>500 mg/liter</td>
</tr>
</tbody>
</table>

For obtaining the linear counting rate-against-discrimination level graph, it was necessary to adjust the electronic gain until the optimum counting conditions were reached. Under these conditions a large fraction of the impulses saturated the non-overloading amplifier. Counting rates were then determined
at different levels of the discrimination voltage. At the lowest discrimination levels used, the background noise due to thermionic emission in the phototube was still considerably less than 1% of the true counting rate.

![Graph showing integral and differential distributions](image)

**Figure 3 a**

Co₆₀: Counting-rate distributions.

Figure 3 shows typical counting-rate distributions found for the radio-nuclides Co₆₀, I¹³¹, Au¹⁹⁸ and P³². The standard deviations of the integral counting rates are not bigger than the plotted points. For Co₆₀ and P³² it is shown how the integral distributions grow progressively more linear as the amplification is increased. At the optimum conditions, the corresponding differential distributions are constant for all practical purposes. The ranges over which the integral counting rate distributions are rectilinear are given in Table II. In this table the lower and higher limits of the linear distributions are expressed as percentages of the extrapolated values. It is seen that the largest extrapolation, arising in the case of Co₆₀, amounts to a correction of 7%. For P³² on the other hand, this reduces to 2%.

It was not possible to decrease the extent of the extrapolations by further increasing the gain. At higher amplifications, a secondary effect set in, namely an abnormal increase in counting rate due to an excess of small amplitude impulses. This is illustrated for Co₆₀ and P³² in figure 4 which, at the lower discrimination levels, shows a sudden departure of the curves from the linear for those cases where the amplification was increased beyond the value necessary for obtaining the straight line distributions.

It was necessary to discriminate completely against these small amplitude impulses during absolute counting since they were not coincident with the
Figure 3 b

$^{131}$I: Counting-rate distributions.

Figure 3 c

$^{198}$Au: Counting-rate distributions.
disintegrations, as was proved by coincidence counting. In \( \beta-\gamma \) coincidence counting, the ratio of the coincidence counting rate to the \( \gamma \) counting rate gives the \( \beta \) efficiency directly. Coincidence determinations were therefore made at different levels of discrimination voltage in the \( \beta \) channel both before and after the sudden increase in counting rate set in. It was found that the above-mentioned ratio did not show the increase as did the \( \beta \) channel counting rate. This is shown for Co\(^{60}\) in figure 5.

The excess of small impulses was not present when background readings were taken with pure scintillator in the counting cell. They are most-probably due to after-pulsing in the multiplier phototube.
Comparisons

Integral counting-rate measurements suitable for extrapolation to zero impulse height, as well as $4\pi \beta-\gamma$ coincidence measurements, were made for each of a number of sources of the radionuclides Co$^{60}$, I$^{131}$ and Au$^{198}$. In order to have a $\beta$ efficiency as high as possible during the coincidence determinations, the discrimination level in the $\beta$ channel was set as low as possible on the straight line before the abnormal increase in counting rate set in. In the $\gamma$ channel, discrimination voltages were set for these measurements at previously determined optimum levels for each nuclide. It has been shown by the author that for every radionuclide a discrimination level can be found in the $\gamma$ channel which renders the coincidence determination practically independent of $\beta$ efficiency over wide variations in $\beta$ efficiency $^{(3)}$.

For $4\pi \beta$ proportional counting of P$^{32}$ sources were prepared in the usual way as thin dry deposits on 50 $\mu$g/cm$^2$ cellulose acetate films made conducting with a coating of vacuum evaporated gold. These sources and the samples taken for the liquid scintillation counting were prepared from the same P$^{32}$ solutions.

Results and Discussion

For each source listed in Tables III, IV and V, the ratios are given of the liquid scintillation extrapolation measurement relative to the $4\pi \beta-\gamma$ coincidence.
Figure 4b

Co\(^{60}\): Excess of impulses at the lowest discrimination levels.

Figure 5

Top: Ratio of coincidence to \(\gamma\) counting-rate as a function of discrimination level.

Bottom: The corresponding \(\beta\) integral counting-rate distribution.
measurement. In Table VI the ratios are given of $4\pi$ liquid scintillation counting measurements relative to $4\pi$ proportional counting measurements on a series of $^{32}P$ solutions. As indicated, some of the measurements are those that were made during international comparisons of standardizations.

Regarding these ratios, the $4\pi\beta-\gamma$ coincidence determinations can confidently be taken as being the more accurate. If in addition the method of $4\pi\beta$ proportional counting of $^{32}P$ is completely relied upon, it seems that there is a measure of energy dependence associated with the $4\pi$ scintillator measurements. (See figure 6.) However, the difference between the liquid scintillation counting and the other methods is more than $1\%$ only in the case of $^{60}Co$ which nuclide represents the lowest energy $\beta$ emitter investigated. In this respect, it is interesting to note the work of SMITH (9) and GUNNINK et al. (4) on the

\[
\text{TABLE III} \\
\text{Co}^{60}: \text{Comparison of Standardizations}
\]

<table>
<thead>
<tr>
<th>Source</th>
<th>Ratio of the $4\pi$ liquid scintillation counting measurement to the $4\pi\beta-\gamma$-coincidence determination</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.9692</td>
</tr>
<tr>
<td>2</td>
<td>0.9867</td>
</tr>
<tr>
<td>3</td>
<td>0.9749</td>
</tr>
<tr>
<td>4</td>
<td>0.9712</td>
</tr>
<tr>
<td>5</td>
<td>0.9805</td>
</tr>
<tr>
<td>6</td>
<td>0.9780</td>
</tr>
<tr>
<td>Mean</td>
<td>0.9768 ± 0.0059</td>
</tr>
</tbody>
</table>

\[
\text{TABLE IV} \\
\text{I}^{131}: \text{Comparison of Standardizations}
\]

<table>
<thead>
<tr>
<th>Source</th>
<th>Ratio of the $4\pi$ liquid scintillation counting measurement to the $4\pi\beta-\gamma$-coincidence determination</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.0010</td>
</tr>
<tr>
<td>2</td>
<td>0.9934</td>
</tr>
<tr>
<td>3</td>
<td>0.9908</td>
</tr>
<tr>
<td>4</td>
<td>0.9873</td>
</tr>
<tr>
<td>5</td>
<td>0.9861</td>
</tr>
<tr>
<td>6</td>
<td>0.9933</td>
</tr>
<tr>
<td>7</td>
<td>0.9950</td>
</tr>
<tr>
<td>Mean</td>
<td>0.9924 ± 0.0046</td>
</tr>
</tbody>
</table>
TABLE V
Au$^{198}$: Comparison of Standardizations *

<table>
<thead>
<tr>
<th>Source</th>
<th>Ratio of the $4\pi$ liquid scintillation counting measurement to the $4\pi$ $\beta$-$\gamma$-coincidence determination</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.9968</td>
</tr>
<tr>
<td>2</td>
<td>0.9978</td>
</tr>
<tr>
<td>3</td>
<td>0.9982</td>
</tr>
<tr>
<td>4</td>
<td>0.9976</td>
</tr>
<tr>
<td>5</td>
<td>0.9996</td>
</tr>
<tr>
<td>6</td>
<td>0.9896</td>
</tr>
<tr>
<td>7</td>
<td>0.9917</td>
</tr>
<tr>
<td>Mean</td>
<td>0.9959 ± 0.0034</td>
</tr>
</tbody>
</table>

* These measurements were made during the international comparisons of the NBS Au$^{198}$ standards, December, 1958.

TABLE VI
P$^{32}$: Excess of impulses at the lower discrimination levels.

<table>
<thead>
<tr>
<th>Solution</th>
<th>Ratio of the $4\pi$ liquid scintillation counting measurement to the $4\pi$ proportional counting measurement</th>
</tr>
</thead>
<tbody>
<tr>
<td>1$^a$</td>
<td>1.0174</td>
</tr>
<tr>
<td>(a)</td>
<td>1.0150</td>
</tr>
<tr>
<td>(b)</td>
<td>1.0161</td>
</tr>
<tr>
<td>2$^a$</td>
<td>1.0049</td>
</tr>
<tr>
<td>(a)</td>
<td>1.0051</td>
</tr>
<tr>
<td>(b)</td>
<td>1.0061</td>
</tr>
<tr>
<td>(c)</td>
<td>1.0088</td>
</tr>
<tr>
<td>3$^{a,b}$</td>
<td>1.0137</td>
</tr>
<tr>
<td>(a)</td>
<td>1.0141</td>
</tr>
<tr>
<td>(b)</td>
<td>1.0087</td>
</tr>
<tr>
<td>(c)</td>
<td>1.0074</td>
</tr>
<tr>
<td>4</td>
<td>1.0067</td>
</tr>
<tr>
<td>5</td>
<td>1.0137</td>
</tr>
<tr>
<td>(a)</td>
<td>1.0141</td>
</tr>
<tr>
<td>(b)</td>
<td>1.0087</td>
</tr>
<tr>
<td>(c)</td>
<td>1.0074</td>
</tr>
<tr>
<td>6</td>
<td>1.0067</td>
</tr>
<tr>
<td>Mean</td>
<td>1.0103 ± 0.0044</td>
</tr>
</tbody>
</table>

* These measurements were made during the international comparisons of the NPL P$^{32}$ standards, June, 1957.

** These measurements were made during the international comparisons of the AECL P$^{32}$ standards, March, 1958.

*** These measurements were made during the international comparisons of the AECL P$^{32}$ standards, April, 1959.
efficiency of the $4\pi$ counting method for Co$^{60}$. These authors report an efficiency of about $90\%$ for sources prepared in the normal way as dry deposits.

A check on the efficiency of the liquid scintillation counting method at a few more energies seems indicated, especially in the lower energy range.

![Figure 6](image)

However, the results reported here already prove that the $4\pi$ liquid scintillation counting method compares favourably with other $4\pi$ counting methods of absolute radioactivity measurement.

REFERENCES


Measurement of $\beta$ Emitters by the Radiated Charge

The method is based on the principle recently described by Failla and his associates:

Two plane electrodes of the same material are mounted parallel to, and close beside, each other in a vacuum container and the source which is to be measured is laid on one of them. The back-scattering on this electrode is offset by the reflections on the other electrode. A magnetic or electrical field makes it possible to eliminate the effect of low-energy secondary electrons.

In the case of magnetic deflection, the apparatus includes an extrapolation chamber and, in the case of electrical deflection, a chamber with a very fine grille.

In the case of Sr$^{90}$-Y$^{90}$ and Pb$^{32}$ the results obtained tally very well with the measurements made with the $4\pi$ counter (better than $\pm 2\%$).

In the case of S$^{35}$ the results are seen to be affected by the atomic number of the substance of which the electrode whereon the source is placed consists; this must therefore be chosen with care.

The theories on back-scattering provide an explanation of these results.

Mesure de l'activité des émetteurs $\beta$ à partir de la charge rayonnée

Le principe de la méthode est celui décrit récemment par Failla et ses collaborateurs:

Deux électrodes planes parallèles, de même substance et très voisines, sont disposées dans une enceinte vidée. Sur une des électrodes est déposée la source à mesurer. La rétrodiffusion sur l'électrode porte-source est compensée par les réflexions sur l'autre électrode. Un champ magnétique ou électrique permet d'éliminer l'effet des électrons secondaires de faible énergie.

Le dispositif expérimental comporte, dans le cas de la déflexion magnétique, une chambre à extrapolation et dans le cas de la déflexion électrique, une chambre à grille très fine.

L'accord avec des mesures effectuées au compteur $4\pi$ est excellent dans le cas du Sr$^{90}$-Y$^{90}$ et du Pb$^{32}$ (mieux que $\pm 2\%$).

Dans le cas du S$^{35}$, les expériences mettent en évidence l'influence du nombre atomique du support de source et en imposent le choix.

Les théories sur la rétrodiffusion permettent d'expliquer ces résultats.

Измерение активности излучателя $\beta$-частиц на основе излучаемого заряда

Принцип метода описан недавно г-ном Пэлла и его сотрудниками:

Два таких электрода, сделанные из одного материала и расположенные параллельно и близко друг от друга, помещаются в вакуумный сосуд. На одном из электродов помещают подлежащий измерению источник. Обратная диффузия на электроде, на котором находится источник, компенсируется
отражением на другом электроде. Магнитное или электрическое поле позволяет устранить действие второстепенных электронов слабой мощности.

Экспериментальный механизм состоит, в случае магнитного отклонения, из экстраполяционной камеры, а в случае электрического отклонения, из камеры с очень тонкой решеткой.

Согласованность с измерениями, осуществляемыми на счетчике 4π, прекрасна для Sr⁹⁰-Y⁹⁰ и P³² (точность более чем 2 процента).

В отношении S³⁵, опыты выявляют влияние атомного номера опорой источника и этим самым определяют его выбор.

Теории об обратной диффузии позволяют объяснить эти результаты.

Medición de la actividad de los emisores β a partir de la carga irradiada

El método se basa en el principio recientemente descrito por Failla y sus colaboradores:

En un recinto en que se ha hecho el vacío se disponen paralelamente y a poca distancia entre sí dos electrodos planos del mismo material. Sobre uno de ellos se deposita la fuente que se ha de medir. La retrodispersión en el electrodo portador de la fuente queda compensada por la reflexión en el otro electrodo. Un campo magnético o eléctrico permite eliminar el efecto de los electrones secundarios de escasa energía.

Cuando la deflexión se efectúa por medios magnéticos, el dispositivo experimental se completa por una cámara de extrapolación y cuando la deflexión es eléctrica, por una cámara de rejilla muy fina.

En el caso del Sr⁹⁰-Y⁹⁰ y del P³², los resultados obtenidos concuerdan satisfactoriamente con los de las mediciones efectuadas con ayuda de un contador 4π (precisión superior a ±2%).

En el caso del S³⁵, las experiencias realizadas ponen de manifiesto la influencia ejercida por el número atómico del soporte de la fuente, lo cual obliga a utilizar determinados soportes.

Los resultados obtenidos pueden explicarse con ayuda de las teorías sobre la retrodispersión.

La méthode de mesure par «perte de charge», bien qu'étudiée depuis 1903 n'a guère été utilisée pour la mesure des radioéléments jusqu'en 1950 (1) (2).

Plusieurs auteurs en ont alors étudié les possibilités et cherché à tourner les difficultés qu'elle présente (3) (4) (5) (6) (7) (8) (9).

En effet, bien que son principe de base en soit très simple, plusieurs phénomènes la rendent d'application difficile.

Le principe de la méthode est le suivant: la source β déposée sur un film conducteur mince est placée dans une enceinte où on fait le vide. Le film, isolé par rapport à l'enceinte, est relié à un dispositif électrométrique.

Pour tout β émis par la source, celle-ci perd une charge de 1,602.10⁻¹⁹ coulombs. Donc une source de 1 millicurie (si à chaque désintégration correspond l'émission d'un β) perdra une charge de 1,602.10⁻¹⁹ × 3,7.10⁷ coulombs/sec, soit: 5,93.10⁻¹² A.
Ainsi la mesure de ce courant permettrait théoriquement d'atteindre l'activité de la source en valeur absolue.

Cependant, plusieurs difficultés doivent d'abord être surmontées. La principale provient du fait que les β arrachent aux atomes de la source ou de l'enceinte, des électrons secondaires de faible énergie (pour la plupart d'énergie inférieure à 40 eV), ce qui se traduit par un accroissement de la charge perdue par la source. Par ailleurs, il faut tenir compte des β réfléchis sur l'enceinte qui reviennent à la source et de l'absorption dans le support de source.

Pour obvier à ces difficultés, CLARK a utilisé la méthode suivante: pour ramener les électrons secondaires d'où ils sont issus, il entoure la source d'une grille portée à une tension négative et pour tenir compte des réflexions des β sur les parois de l'enceinte, il remplace la grille par un écran de mêmes dimensions, puis par une grille et un écran de plus grandes dimensions (figure 1 a).

Plus récemment, FAILLA et ROSSI ont proposé une méthode différente (8). La source est déposée au centre d'une plaque conductrice d'épaisseur supérieure au parcours des β. Une plaque identique est placée parallèlement, à une distance de quelques mm. Entre les deux plaques, une grille très fine, portée à un potentiel négatif repousse les électrons secondaires sur les électrodes d'où ils sont issus. Du fait de leur symétrie, la perte et le gain en électrons des deux plaques sont
les mêmes et le courant électronique quittant la plaque porte-source est égal à la moitié du courant qui serait produit par le départ de tous les β (figure 1 b). Une variante de la méthode consiste à utiliser au lieu du champ électrique créé par la grille, un champ magnétique.

Dans cette étude, encore en cours, nous avons utilisé la méthode de Failla et Rossi, qui nous a paru séduisante par sa simplicité, avec l'objectif d'en explorer les possibilités et les limitations d'emploi et d'en rechercher les conditions les plus pratiques d'utilisation.

**Théorie de la Méthode**

Considérons deux plaques parallèles identiques de dimensions infinies. L'une sera l'électrode porte-source; nous appellerons l'autre contre-électrode. Faisons abstraction d'abord des électrons secondaires.

Soit \( N \) le nombre des \( \beta \) émis par la source par seconde en \( 4\pi \), soit \( R \) le coefficient de rétrodiffusion de l'électrode vis-à-vis des \( \beta \) (coefficient qui tient compte de tous les rétrodiffusés émis dans l'angle solide \( 2\pi \)).

Lors des réflexions successives, même si elles s'accompagnent d'une dégradation d'énergie, \( R \) sera sensiblement constant puisqu'il ne dépend que du \( n^\circ \) atomique de la substance.

Calculons alors le nombre de \( \beta \) qui quittent l'électrode porte-source par seconde.

Si cette électrode était seule, le nombre de \( \beta \) émis serait \( \frac{N}{2} (1 + R) \). La contre-électrode en réfléchit à son tour \( \frac{N}{2} (1 + R) R \), dont \( \frac{N}{2} (1 + R) R^2 \) seront réémis par l'électrode porte-source, etc. En définitive, le nombre d'électrons perdus par l'électrode porte-source sera:

\[
\begin{align*}
n &= \frac{N}{2} (1 + R) - \frac{N}{2} (1 + R) R + \frac{N}{2} (1 + R) R^2 - \frac{N}{2} (1 + R) R^3 + \ldots \\
&= \frac{N}{2} (1 + R) (1 - R) (1 + R^2 + R^4 + \ldots) \\
&= \frac{1}{1 - R^2} \quad \text{et} \quad n = \frac{N}{2}.
\end{align*}
\]

Pour se rapprocher des conditions théoriques, il faut évidemment que l'angle solide sous lequel est vue la contre-électrode de la source soit le plus petit possible, mais les dimensions minimum à adopter sont difficilement calculables car la probabilité de réflexions multiples est assez grande.

Cette condition sera difficile à respecter dans le cas de la chambre à déflexion magnétique, car le champ doit être uniforme dans tout l'espace interélectrode et d'intensité d'autant plus grande que la distance interélectrode sera plus faible.

Dans le cas de la chambre à grille se présente une difficulté d'un autre ordre: il faut que la grille soit suffisamment fine pour ne pas créer d'électrons secondaires qu'elle repousserait.
Appareillage utilisé

a) Chambre à grille

Les deux électrodes ont 11 cm de diamètre et sont distantes de 3 mm. Entre elles est tendue une grille faite de fils fins de nylon recouverts d'aquadag; la masse superficielle moyenne de la grille est d'environ 0.1 mg/cm².

Les deux électrodes sont permutables de telle sorte qu'on peut mesurer soit la perte de charge de la source, soit la collection des électrons.

b) Chambre à champ magnétique

Pour tourner la difficulté signalée précédemment, nous avions utilisé une chambre à extrapolation: la distance interélectrode est rendue variable sans qu'il soit nécessaire de «casser» le vide.

Les électrodes ont un diamètre de 5.5 cm seulement (figure 2). Cette chambre permet ainsi de tracer la courbe du courant mesuré en fonction de la distance, courbe que l'on extrapole à zéro. Ici encore, on a possibilité par permutation des électrodes de mesurer la perte de charge ou la collection électronique.

Ces chambres sont reliées à un banc à vide permettant d'atteindre un vide de $10^{-5}$ mm de mercure, et connectées à un dispositif électrométrique comportant un électromètre Lindemann et un condensateur étalon (méthode de zéro de Townsend) (Figure 3).
Étude avec divers radioéléments

1. Source de Sr⁰⁰ — Y⁰⁰

Nous avons fait des essais systématiques avec une source déposée, avec mouillant, au centre d'un disque d'aluminium.

L'activité de la source, d'après la mesure au compteur 4π était de 114,8 μc ± 3%.

Figure 3

a) Chambre à champ électrique

Nous avons observé d'abord que les résultats ne sont bien reproductibles que si la tension d'alimentation de la grille a une dérive très faible. Le calcul montre d'ailleurs que dans nos conditions expérimentales (capacité grille — collectrice 50 μF, courant mesuré de l'ordre de 3,5 × 10⁻¹³ A, tension grille de 0 à 250 V) pour que l'erreur sur le courant mesuré due à cette dérive soit inférieure à 0,5%, il faut une dérive horaire de la tension inférieure à 0,5%/h. En particulier, l'alimentation par piles est à prohiber.

La mesure s'effectue en mesurant le courant pour des valeurs négatives croissantes de la tension grille et en extrapolant la 2e partie de la courbe obtenue (déflexion des β) à la tension zéro.

Nous avons effectué pour la même source la mesure de la perte de charge et de la collection électronique. Les deux résultats sont identiques et
correspondent à une activité de 116,2 μc. Cependant, le fait que les deux résultats soient identiques ne prouve pas obligatoirement qu’il n’y a pas «échappement des β» sur les côtés des électrodes, car ceux-ci pourraient-être compensés par les électrons secondaires produits par la grille.

Finalement, c’est la comparaison avec la chambre à champ magnétique qui nous permettra de tester la valeur de ce résultat.

b) 

**Chambre à champ magnétique**

Nous opérons de la manière suivante.

Pour chaque valeur d de la distance interélectrode, nous traçons la courbe du courant en fonction de la valeur du champ magnétique. La courbe peut être décomposée en deux parties: déflexion des électrons secondaires (courbe à forte pente), puis déflexion des β les plus mous. On extrapole à zéro cette 2e partie de la courbe.

Pratiquement dans notre dispositif expérimental, on observe que le point de raccordement des deux courbes se situe vers une valeur de H telle que H d 50 gauss-cm.

On porte alors les valeurs extrapolées du courant en perte de charge et en collection électronique en fonction de la distance interélectrode. Les deux courbes obtenues concourent, comme on pouvait s’y attendre au même point pour d = 0. Mais ce qui n’était pas évident à priori, et que nous n’avons d’ailleurs pas réussi à démontrer théoriquement, la moyenne des deux courants pour chaque valeur de d se situe très sensiblement sur une horizontale (Figure 4).

Cette propriété, qui s’est vérifiée sur toutes les autres sources passées à la chambre permet d’obtenir une très bonne précision sur l’extrapolation à la distance zéro.

Figure 4
La valeur obtenue par cette méthode était de 115,5 μc, en bon accord avec la valeur donnée par la chambre à grille. Ces résultats doivent subir une légère correction du fait du phénomène "d'ionisation interne".

En effet, lors de la désintégration β, une partie de l'énergie de la transition peut se retrouver sur un électron du cortège qui se trouve alors éjecté de l'atome. Le réarrangement du cortège est accompagné aussi de l'émission de photons et d'électrons Auger.

En extrapolant au Sr-Y les résultats expérimentaux les plus récents, on trouve que cette correction est de l'ordre de 0,3% ce qui, pour les deux chambres donne respectivement 115,8 et 115,1, en très bon accord avec la valeur de 114,8 obtenue au compteur 4π.

2. Sources de P³²

La correction pour l'ionisation interne est ici plus importante: environ 1,6%.

Compté-tenu de cette correction, nos résultats ont été les suivants: 89,6 μc pour une source de 88,6 μc ± 3% d'après le compteur 4π et 51,6 et 51,1 μc pour deux sources de 50,5 μc ± 3%, donc en bon accord.

3. Sources de S³⁵

Des résultats peu encourageants au point de vue des dépôts nous ont montré qu'il y a intérêt à déposer la solution sur une feuille métallique laminée plutôt que sur un support usiné même bien poli. En effet, la moindre aspérité ou poussière provoque des dégradations cristallines malgré la présence de l'agent mouillant. Cependant, malgré ces précautions prises, et à cause de l'activité dont nous avions besoin pour ces expériences, l'épaisseur des plus gros cristaux que nous avons obtenus était de l'ordre de 0,5 à 1 mg/cm², ce qui correspondrait à une absorption auto de l'ordre de 5% pour les rayons directs. Mais pour les rayons rétrodiffusés, l'absorption est plus importante, et on ne peut plus considérer alors que l'on a compensation des rétrodiffusés entre les deux électrodes.

Par ailleurs, on doit s'attendre à ce que cette absorption soit spécialement importante dans le cas de dépôts sur feuille de faible numéro atomique (par exemple l'aluminium), car la rétrodiffusion s'accompagne alors d'une importante dégradation d'énergie.

Nos résultats avec des dépôts sur feuille d'aluminium sont en moyenne 10% inférieurs à ceux du 4π.

Pour éviter la dégradation d'énergie des rétrodiffusés, nous avons essayé des dépôts sur feuille d'or. Mais ici intervient un phénomène tout aussi gênant: le numéro atomique moyen du dépôt est voisin de celui de l'aluminium. Or une partie des rétrodiffusés le sont par la source elle-même, dont le coefficient de rétrodiffusion est inférieur à celui de l'or. Donc dans ce cas encore, si la source n'est pas sans matière, on n'aura plus compensation des rétrodiffusés. Pour des dépôts sur feuille d'or, nos résultats étaient en moyenne 15% inférieurs à ceux du 4π.

Malgré ces résultats négatifs, nous pensons persévérer en essayant d'autres techniques de préparation de sources.
Conclusion

Nous pensons que la méthode est viable pour de nombreux radioéléments émetteurs β ou β, γ à condition de disposer de solutions avec très peu d'entraineur. On ne peut guère faire de calcul d'autoabsorption, aussi le point délicat de la méthode réside dans la préparation des sources pour obtenir les cristaux les plus petits possibles.

Il nous semble que pour la mesure des émetteurs β d'énergie maximum supérieurs à 0.3 MeV, il est possible de construire un appareil d'un maniement simple susceptible de donner une bonne précision.

En particulier, contrairement à ce qui est signalé dans la littérature, il ne nous semble pas nécessaire d'utiliser pour la mesure du courant une méthode de zéro (qui est cependant à recommander pour des raisons de précision). En effet, l'objection avancée à ce propos est que la différence de potentiel entre la source et l'enceinte pourrait prendre des valeurs élevées. Or il n'en est rien avec un amplificateur à courant continu puisque sa résistance d'entrée permet d'écouler les charges. Il faut cependant que la différence de potentiel entre la collectrice et l'enceinte reste très faible pour éviter de collecter les électrons secondaires; or cette condition est réalisée avec les amplificateurs à contre-réaction totale.

La seule précaution à prendre est de soustraire la lampe électromètre à l'influence du champ magnétique car l'application de celui-ci change le zéro de l'amplificateur.

Les essais que nous avons effectués avec un tel amplificateur nous ont donné un excellent accord avec la méthode de Townsend.

Compte-tenu de nos essais, nous pensons réaliser un appareil modifié dont les caractéristiques seront les suivantes:

Une chambre à champ magnétique dont la distance interélectrode sera de l'ordre de 5 mm. De la sorte, le champ nécessaire sera faible et l'extrapolation au champ zéro facile. Chaque électrode pourra être couverte par une feuille métallique tendue à l'aide d'une bague-ressort.

Nous utiliserons la propriété que nous avons signalée précédemment (à savoir que la somme des valeurs absolues du courant de perte de charge de l'électrode porte-source et du courant électronique de la contre-électrode donne la charge totale rayonnée par la source en 4\pi, indépendamment de la distance interélectrode) de la manière suivante.


La seule courbe à tracer serait celle du courant en fonction du champ magnétique, ce qui serait l'équivalent pour l'appareil du tracé des paliers de compteur.

En pratique, un tel appareil pourrait rendre des services pour le contrôle des solutions mères, le seuil d'activité mesurable avec bonne précision étant de l'ordre de 20\muC.
(2) RUTHERFORD, E. et al., Radiations from Radioactive substances, p. 387 (1930).
(8) FAILLA, G., ROSSI, HH., *NYO* 4523 (1953).
II. 2. 10. THE ABSOLUTE MEASUREMENT OF THE RADIOACTIVITY OF GASES

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Abstract

After a survey of the present state of development of methods for the absolute measurement of the activity (decay rate) of radioactive substances, the measurement of radioactivity in gases (internal gas counting) is treated in more detail. In particular, methods for finding and correcting mistakes which arise in this method of measuring are suggested. The absolute accuracy of measurements obtained by the isotope laboratory with the correction methods referred to is at present about 2%; it should be further improved in the near future.
The isotope laboratory of the “Kernreaktor Bau- und Betriebs-Gesellschaft”, Karlsruhe, which was a member of the Max Planck Society in Göttingen up to 1957, has for some time been carrying on work in the field of the absolute measurement of radioactive substances. A number of years ago the measurement programme was extended to include the absolute measurement of radioactive gases. The purpose of the investigation was to study as many as possible of the factors that might impair accuracy of measurement and to determine their influence on the result. A particular aim was to determine the necessary corrections with the maximum degree of accuracy.

All tests and measurements were carried out in our laboratory by Mr. Spernol, now with the Nuclear Research Centre at Mol, Belgium.

After critical examination of all known procedures for the absolute measurement of radioactive gases, we came to the conclusion that the following method was most appropriate for the purpose of our investigation. The radioactive substance to be measured is introduced in gaseous form, usually mixed with a suitable counter gas, into a counter tube and the disintegration rate is measured. In general, the direct introduction of $\beta$ emitters into the counter tube as gases ensures 100% efficiency of the counter. A particular advantage of this method is that it prevents any absorption of $\beta$ radiation, i.e. including self-absorption by the radiation source, so that the procedure is suitable even with low $\beta$ irradiators, such as tritium and $^{14}C$.

However there are a number of systematic error sources, and the study of these has been the main objective of our investigation. The most important sources of error are the following:

1. In general, the radioactive initial substance is not a gas, but either a solid or a liquid. In the necessary gasification, which is usually carried out by chemical methods, an isotope effect may, under certain circumstances, be produced if conversion is incomplete.

2. By reason of the chemical structure of the gas to be examined, it will in the majority of cases be necessary to determine the disintegration rate in a proportional counter. If the electronic arrangement is inadequate and the discriminator voltage is high, low-energy $\beta$ particles may escape the count.

3. In practice it will hardly be possible so to arrange the geometrical form of the counter tube that its effective volume is identical with its actual volume.

4. Owing to the adsorption or absorption of radioactive material in the walls of the counter, radioactive substance may escape the count, while, on the other hand, radioactive gas from previous measurements may be diffused back into the counter tube, giving rise to a memory effect.

A considerable number of methods have been described for the conversion of tritium-marked water into gases containing tritium, for example hydrogen, or methane, and of barium carbonate marked with $^{14}C$ into gases containing $^{14}C$, for example carbon dioxide, or methane. We are also familiar with direct methods of converting complicated marked substances into the corresponding measurable gases without passing through the intermediate stage of water or barium carbonate. For the measurement of $^{14}C$ we selected the method often quoted in chemical literature of preparing CO$_2$ from barium carbonate and sulphuric acid, while for the measurement of tritium we developed a precise macro-method out of Dubbs's micro-method, by which water is reduced at 400° C, using zinc as a deoxidizer. Both methods now give a counting yield of 99% to 101%, and repro-
ducibility to 99.8% in similar experiment series. The quantitative yield of tritium or C\textsuperscript{14} ensures that no isotope effects occur in the preparation of the gas.

No difficulties are met with in counting the hydrogen or carbon dioxide prepared. Methane, ethylene, butane, etc., are particularly well suited for use as counter gases to be mixed with the gas samples. The only electronic problem, especially in the measurement of tritium, lies in the provision of a sensitive non-overloading amplifier. We have had satisfactory experience with the Higinbotham ORNL amplifier and with the Tracerlab amplifier. With the plateaux as measured by means of a Tracerlab amplifier the counting rate can be read off to an accuracy of 0.3% with a carbon dioxide/methane filling, and 0.5% with a tritium/methane filling.

The correction of the counter-tube efficiency to compensate for the low-energy β particles not counted depends on the discriminator voltage selected. If the discriminator voltage can be converted into corresponding particle energies measured in eV, the counting loss can be determined on the basis of the known integral β-ray spectra. The discriminator voltages were calibrated by recording the differential spectra of Fe\textsuperscript{55} with a K-Line at approx. 6 kV and of Ar\textsuperscript{37} with an L-line at 280 V. The discriminator energy selected for tritium measurements was 180 eV; that for C\textsuperscript{14} was approximately 1000 eV. The resulting counting losses were approximately 2% for tritium and approximately 1% for C\textsuperscript{14}.

Surface effects in the counter tube can be divided into those occurring only at the ends of the counter tube and independent of the counter-tube length, and those occurring on the counter-tube walls and largely proportional to the length of the counter tube. The end effects are produced by disturbance in the potential at the end of the counter tube and by irradiation from the insensitive volume at the end of the counter tube into the effective volume. These end effects can easily be eliminated by the simultaneous measurement of the counter gases in two or more counter tubes of different lengths but otherwise identical construction. The difference between the counting rates in the individual counter tubes corresponds exactly to the difference in volume of the counter tubes, so that it is possible to establish the relationship between the absolute counting rate and the unit of volume. We investigated the end effects by making measurements with four counter tubes of different lengths, at different counter-gas pressures, and by recording the potential in models. The results show that in normal counter-tube construction the end disturbance does not penetrate more than 1—2 diameters into the effective volume, and that it is possible to build counter tubes for β irradiators of not too low energy, e. g. Kr\textsuperscript{85}, in which, at certain pressures, the counting losses and the counting gain exactly balance one another out through the end effects. In one of our standard counter tubes, 50 cm in length, 4 cm in diameter, with shield tubes 3 cm in length, and 60 cm Hg pressure of gas filling, the total end-effect correction was -5% with tritium and approx. -3% with C\textsuperscript{14}.

By means of measurements made with counters of different diameters we endeavoured to discover the wall effects. For C\textsuperscript{14} it was found that the wall effect was proportional to the reciprocal of the diameter, i. e. to the wall area per unit of volume. With a counter-tube diameter of, say, 4 cm, the necessary correction is approx. 2.5%. Before one can set up correction formulae for the wall effects from which, under certain circumstances, it is
possible to determine the most favourable measuring conditions, one must investigate the individual wall effects separately. Wall effects are due to the penetration of electrons into the wall of the counter-tube, the fraction of energy lost in the counter gas being so small that it escapes detection. Normally, the purely geometric effect can easily be calculated. For this purpose one must be able to state the mean distance — dependent on the counter-gas pressure — over which the electrons in the counter tube lose their discriminator bias. Only from a layer of this thickness adjoining the wall can electrons penetrate into the wall uncounted. A simple geometrical calculation shows that an average of one quarter of all electrons from this layer are not counted.

This purely geometrical effect is overlaid by the back-scattering of electrons and by phenomena of absorption, adsorption, and chemisorption. We need not take secondary electrons into consideration because of their low energy. Under our test conditions, counting losses arising from these absorption phenomena are no greater than approx. 1% with carbon dioxide/methane mixtures, even when the radioactive and inactive carbon dioxide are mixed in the proportion 1:100. In tritium measurements, however, the counting losses arising from these surface effects may attain a considerable percentage figure, so that it proved necessary to examine these effects more closely. We calculated the absorption arising from diffusion into the walls by measuring against time the drop in the counting rate after filling of the counter tube, and by measuring the activity diffused back from the wall after inactive refilling of the counter tube. From the solution of the diffusion equation for both directions of diffusion it is possible to establish exact formulae for the time dependence of the absorption correction. Our measurements indicated good correspondence between time dependence as calculated theoretically and as established by experiment.

The most important result of these investigations is that one can always avoid having to make an absorption correction of more than 0.1% if the counting is done soon enough after the filling of the counter tube — i.e. within a few minutes.

This is not possible in correcting for adsorption, as in adsorption a proportion of the counter gas adheres to the walls almost immediately. It is also possible to give a theoretical formula for adsorption, and the constant of this formula can be calculated from measurements made with counters of different diameters or by variation of the inactive hydrogen content of the counter gas. The results of these measurements show strong fluctuations. For our standard tritium test conditions we obtained an adsorption of 1—4%.

The basis of chemisorption is a strong bonding, and sometimes exchange, of the active substance in the counter-tube wall, taking effect as a residual contamination in the counter-tube. In $^14C$ measurements we found a residual contamination persisting for several years, for example on plastic insulators of a certain material. All chemical surface effects are dependent to a very large extent on the nature of the material in question. We therefore conducted a series of experiments on the dependence of surface effects on the nature of the material. Our results showed particularly low absorption effects with copper, no difference being noted between steel, aluminium and brass. Measurements with tritium have so far not yielded a clear picture, but steel and copper seem to be particularly well suited for use with this element.
We now hope that, bearing in mind all the sources of error mentioned in this paper and applying all the corrections calculated, we can carry out absolute measurements with tritium and C\textsuperscript{14} to an accuracy of approx. 2—3\%. As in all absolute measurements, however, we are not sure whether other systematic errors, of which we are unaware, may not be considerably influencing the measurements. It is our belief that we might perhaps be able to discover such errors by comparing results with other laboratories. We should therefore be very glad to be given the opportunity of making comparative measurements with tritium and C\textsuperscript{14} in cooperation with other laboratories.
II. 2. 11. RECENT WORK WITH COMPENSATED INTERNAL GAS COUNTERS FOR THE STANDARDIZATION OF GASEOUS RADIONUCLIDES

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Abstract

The criteria for compensation in internal gas counters and recent applications of such counters at the National Bureau of Standards will be reviewed.

Travaux récents d'étalonnage de radionuclides gazeux, effectués au moyen de compteurs compensés à gaz de comptage interne

L'auteur examine les critères régissant la compensation dans les compteurs à gaz interne et les applications récentes de ces compteurs au National Bureau of Standards des Etats-Unis.

Последние работы со счетчиками с компрессированным внутренним газом для стандартизации радионизотопов в виде газа

Будут обсуждены критерии для компенсации в счетчиках с веденным внутрь газом, а также последние применения таких счетчиков в Национальном бюро стандартов.

Recientes investigaciones con contadores compensados de gas interno para la calibración de radionúclidos gaseosos

En la memoria se examinan los criterios que se siguen para la compensación de los contadores de gas interno y las recientes aplicaciones de estos aparatos en el National Bureau of Standards de los Estados Unidos.

This brief paper is merely intended as a review of recent work with our internal gas-counting systems with particular reference to some new results that may be of interest.

Unfortunately most of the work with our gas counters has been described in short papers such as this one but we hope soon to collect all the information about them together for formal publication.

I would like to show a few slides which give details of the counters and of the associated gas-handling equipment.

The principle of compensation depends on the distribution of electric intensity between the anode and cathode near the ends of both the long and short counters being the same, and of there being a region in the center of each counter where the electric intensities are both equal and uniform. If a shorter counter were too short then one might get a distribution such as is shown by the third curve in figure 1 in which case compensation would not be achieved. We have worked therefore with one long and two shorter counters of different length (a "medium" and a "short" counter), in order to assure ourselves that the short counter was not too short. We have also constructed the counters from stainless steel and from copper. The stainless
steel counters are illustrated in figure 2 while both sets of counters are shown in figures 3 and 4. Great care was taken to make the end insulating structures as precisely the same as possible.

Figure 5 shows the iron shield in which the counters are housed and the auxiliary gas-handling equipment. Behind the shield is an automatic Toeppler pump for completely mixing the radioactive gas specimen and the counting gases.
Figure 5
COPPER COMPENSATED GAS COUNTERS
PRESSURE: 750.92 X 10^3 DYNES/CM²

Figure 6
Figure 6 and 7 show plateaux which were obtained counting C¹⁴ in the form of carbon dioxide in the proportional region with methane as the counting gas in both sets of counters. You will note that all the counters exhibit a slight slope in their plateaux but that the difference “L-S” is practically flat.

Furthermore “L-S” in each case comprises two sets of points the one being simply the difference in the counting rates, \( N_L - N_S \), of the long and the short counters while the other is

\[
(N_L - N_M) \times \frac{V_L - V_S}{V_L - V_M},
\]

where \((V_L-V_S)/(V_L-V_M)\) is simply the factor that involves the differences in volumes of the long and short and long and medium counters. The good agreement of the “L-S” points shows that the short counters are not too short for compensation.

One important effect that has to be remembered in internal gas counting is the so-called “wall effect”, which may possibly arise from β particles striking the counter wall without having created enough ion pairs to give a pulse. This wall effect will be a function of the pressure and can be eliminated by plotting counting rate against the reciprocal of the pressure and extrapolating to zero inverse pressure. Two such extrapolations which were publi-
shed by Dr. Seliger and myself in the Proceedings of the Second Geneva Conference are shown in figure 8. These are with Kr\(^{85}\) but it will be noted that while there appears to be a wall effect with the stainless steel counters there is little or none for the set of copper counters. I had been puzzled by the fact that both these counters and the copper counters that I had used at Chalk River exhibited a far smaller wall effect than would be expected from the anticipated number of ion pairs per centimeter. I mentioned this fact at the Easton conference of October 1957 and both Dr. Borkowski and Dr. Grummitt suggested that the effect might be reduced by both back-scattering and by secondary emission. From figure 8 it would appear that the mechanism may be chiefly secondary emission from the copper counters, possibly from a thin copper oxide layer, but that there is no such compensating effect in the stainless steel counters.

There is one other quite interesting side to the question of compensation. The existence of a slope to the plateau is probably due both to small pulses in the body of the counter which, with greater multiplication with increased voltage, become detectable, and also to the greater number of pulses that are detected as the active volume of the counter expands into the ends of the counter with increasing voltage. The latter effect is susceptible to compensation but the former, due to counts in the body of the counter, is not.

Thus by taking differences between counters of different length we should be able to flatten the plateaux which we obtain as a function of voltage at constant discrimination and also as a function of discrimination at constant
voltage. It may not be reasonable to expect completely flat plateaux with voltage, however, unless it happens accidentally due to some effect such as an increase in dead-time in the proportional counter with increasing voltage.

**Long Copper Counters Minus Short Copper Counter**

*Message Registers × 256 Per Minute*

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**Figure 9**

In figure 9 is shown a matrix of results which were obtained counting Kr\(^{85}\) in the proportional region in the long and short copper counters. The negative voltages on the cathode are shown in the left-hand column and the discriminator voltages horizontally across the top. (To preserve good electrical field conditions the anode wire is at ground potential.) The top row of figures in the matrix gives the message register (on a scale of 256) per minute for the long counter \((N_L)\); the second row the corresponding figures for the short counter \((N_S)\); and the third the difference \(N_L - N_S\). It is seen that within the limits of the border in the figure the values of \(N_L - N_S\) are very reasonably constant. In most cases, however, there is still observed a slight residual increase of counting rate with cathode voltage but the agreement between \(N_L - N_S\) and \((N_L - N_M)/(V_L - V_S)/(V_L - V_M)\) is usually within 1\% or less.

Recently we have been attempting to make a re-determination of the half-life of C\(^{14}\). The calculations for the gram molecule of gas in the counters and for the various counting rates are fairly tedious and so we have established the necessary coding to do this and other calculations of results from the gas counters on the IBM 704 computer.
Ditro 41 cm copper counters 3.0—5.0 disc
Gram moles $\text{CO}_2 = 43.7934 \text{ E—09}$
Pressure $\text{CO}_2 = 5.476 \text{ cm Hg}$
Density $\text{Hg} = 13.5371 \text{ g/ml}$
Kelvin temperature 296.72
$A = 0.36400 \ E - 13$
$B = 42.670$
$V = 0.0149 \text{ ml}$
Gram moles Argon + $\text{ETOH} = 75.585 \text{ cm Hg}$
Pressure Argon + $\text{ETOH} = 75.585 \text{ cm Hg}$
Density $\text{Hg} = 13.5383 \text{ g/ml}$
Kelvin temperature 297.14
$A = 0.22830 \ E - 13$
$B = 42.780$
$V = 11858.000 \text{ ml}$
Gram moles $\text{CO}_2$ in counter 25.74279 $\text{ E—11}$

Gram moles gas = 28.37449 $\text{ E—04}$
Pressure = 41.163 cm Hg
Density $\text{Hg} = 13.5387 \text{ g/ml}$
Kelvin temperature = 298.24

$A = 0.2283000 \ E - 13$
$B = 42.780$
$V = 128.6820 \text{ ml}$
Counter scale factor = 256.
Instrument factor = 1.24990
Dead time = 5.21000 E—05 sec
Length of count = 1.00 min
Number of voltages used = 9

<table>
<thead>
<tr>
<th>Voltage</th>
<th>Long reg</th>
<th>C/S</th>
<th>NL</th>
<th>Med reg</th>
<th>C/S</th>
<th>NM</th>
<th>short reg</th>
<th>C/S</th>
<th>NS</th>
<th>(NL-NM) X</th>
<th>IF</th>
<th>(NL-NS)</th>
</tr>
</thead>
<tbody>
<tr>
<td>50 volts</td>
<td>2900</td>
<td>137.4</td>
<td>586.240</td>
<td>604.710</td>
<td>67.6</td>
<td>288.427</td>
<td>292.827</td>
<td>50.8</td>
<td>216.747</td>
<td>219.222</td>
<td>389.822</td>
<td>385.467</td>
</tr>
<tr>
<td>3000</td>
<td>138.8</td>
<td>592.213</td>
<td>611.067</td>
<td>69.0</td>
<td>294.400</td>
<td>298.886</td>
<td>51.2</td>
<td>218.453</td>
<td>220.968</td>
<td>390.071</td>
<td>390.099</td>
<td></td>
</tr>
<tr>
<td>3100</td>
<td>138.3</td>
<td>590.080</td>
<td>608.796</td>
<td>69.2</td>
<td>295.253</td>
<td>299.866</td>
<td>52.5</td>
<td>224.000</td>
<td>226.645</td>
<td>386.132</td>
<td>382.151</td>
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<tr>
<td>3200</td>
<td>139.3</td>
<td>594.347</td>
<td>613.339</td>
<td>69.5</td>
<td>296.533</td>
<td>301.186</td>
<td>52.2</td>
<td>222.720</td>
<td>225.335</td>
<td>390.159</td>
<td>388.004</td>
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<tr>
<td>3300</td>
<td>140.6</td>
<td>599.893</td>
<td>619.248</td>
<td>70.8</td>
<td>302.080</td>
<td>306.910</td>
<td>53.2</td>
<td>226.987</td>
<td>229.703</td>
<td>390.390</td>
<td>389.544</td>
<td></td>
</tr>
</tbody>
</table>

Figure 10
Figure 10 shows the computations for a number of plateau voltages at 30 and 50 volts discrimination. The final values for both $N_L - N_S$ and for $(N_L - N_M) (V_L - V_S) / (V_L - V_M)$ are shown. The latter is in the column $(N_L - N_M) \times IF$, i.e., instrument factor. The constants $A$ and $B$ are the van der Waals' constants for methane, not argon and ethyl alcohol as labelled in this case.

The IBM 704 computer prints out in about four minutes results that otherwise take some four days of manual computations to obtain.
II. 2. 12. О НЕКОТОРЫХ МЕТОДАХ ИЗМЕРЕНИЯ Р-АКТИВНЫХ ГАЗОВ

В. В. БОЧКАРЕВ, В. А. БАЖЕНОВ
СССР

Abstract

On some methods of the measurement of the $\beta$-active gases

A number of methods for the absolute measurements of $\beta$-active gases by means of special devices, in which gas discharge counters are used, and also some methods of relative and absolute measurements of the activities of readily volatile liquids, are discussed.

Formulas are given for the calculation of the theoretical counting rate: of (1) a cylindrical counter placed in a co-axial vessel, taking into account the specific activity of the filling gas; (2) a device consisting of an end-window counter introduced into a cylindrical vessel.

The methods of more accurate measurements of $\beta$ active gases by means of the gas flow counter, as well as the methods of relative and absolute measurements of the activity of some $\beta$-active liquids through their vapours, are described.

Quelques méthodes de mesure des gaz $\beta$ actifs

Les auteurs discutent un certain nombre de méthodes de mesure absolue de gaz $\beta$ actifs à l'aide de dispositifs spéciaux utilisant des compteurs à décharge dans le gaz, ainsi que plusieurs méthodes de mesure relative et absolue de l'activité de liquides volatils.

On établit les formules de calcul du taux de comptage théorique 1° d'un compteur cylindrique placé dans une enceinte coaxiale, compte tenu de l'activité spécifique du gaz de remplissage du compteur; 2° d'un dispositif constitué par un compteur à fenêtre en bout, plongé dans l'enceinte cylindrique.

On décrit enfin des méthodes de mesure précise de gaz $\beta$ actifs à l'aide du compteur à courant gazeux, de même que des méthodes de mesure relative et absolue de certains liquides $\beta$ actifs, par l’intermédiaire de leurs vapeurs.

Методы абсолютных измерений $\beta$-активных газов

Рассматривается ряд методов абсолютных измерений $\beta$-активных газов с помощью специальных устройств, в которых используются газоразрядные счетчики, а также некоторые методы относительных и абсолютных измерений активности легколетучих жидкостей.

Даны формулы для расчета ожидаемой скорости счета счетчика в зависимости от удельной активности исследуемого газа для устройства, состоящего из цилиндрического счетчика, размещенного на оси цилиндрического сосуда, а также для устройства, состоящего из торцевого счетчика, смотрящего в цилиндрический сосуд.

Описаны методы более точных измерений $\beta$-активных газов с помощью счетчиков внутреннего заполнения, а также методы относительных и абсолютных измерений активности некоторых $\beta$-активных жидкостей по их парам.
Métodos de medición absoluta de la actividad de gases β activos

Los autores estudian diferentes métodos de medición absoluta de la actividad de gases β activos con ayuda de dispositivos especiales en que se utilizan contadores de descarga gaseosa; examinan también algunos procedimientos de medición absoluta y relativa de la actividad de líquidos volátiles.

Dan fórmulas para calcular los índices de recuento teóricos: 1° de un contador cilíndrico colocado en un recinto coaxial, teniendo en cuenta la actividad específica del gas contenido en el contador. 2° de un dispositivo constituido por un contador de ventana terminal introducido en un recinto cilíndrico.

Por último, los autores describen procedimientos para medir con precisión la actividad de gases β activos empleando contadores de corriente gaseosa, así como métodos de medición absoluta y relativa de la actividad de algunos líquidos emisores de rayos β a partir de sus vapores.

Проведение абсолютных измерений активности газообразных β-излучателей связано с большими трудностями, чем для твердых растворимых соединений. Однако, в настоящее время разработан ряд сравнительно простых методов измерения радиоактивных газов. Некоторые из них будут рассмотрены ниже. Это не прецизионные методы, но как нам кажется — они представляют интерес ввиду их доступности.

Абсолютные измерения становятся осуществимыми, коль скоро мы находим возможность теоретического определения, в условиях данного
опыта, величины \( k \) в формуле \( a = kN \), т. е. определения коэффициента пропорциональности, связывающего показания измерительного устройства \( N \) с активностью исследуемого препарата \( a \).

Если в цилиндрическом сосуде по его оси расположить тонкостенный цилиндрический счетчик \( \beta \)-частиц, то получится простое устройство для измерения активности газа. Рецепт определения \( k \) для данной геометрии был дан в 1948 г. (1) при следующих условиях и допущениях: концентрация газа считалась одинаковой по всему фиксированному стенками камеры объему; поглощение \( \beta \)-частиц в находящемся в камере газа и рассеяние от стенок камеры не учитывалось, размеры счетчика и камеры связывались условием \( H - h \geq 2r \), где \( r \) — радиус счетчика, \( H \) — полувысота камеры и \( h \) — половина длины рабочего объема счетчика (см. рисунок 1). При этом была получена довольно простая формула, связывающая число частиц, падающих на боковую поверхность счетчика \( N_0 \) с числом распадов происходящих в 1 см\(^3\) объема камеры в секунду \( n_0 \), с тем большей точностью, чем больше отношение радиусов камеры и счетчика \( R/r \). Причем для \( R/r = 5 \) ошибка не превышает 6,7%.

Для того, чтобы получить число частиц, регистрируемых счетчиком в секунду, необходимо сделать обычными методами поправку на поглощение в стенках счетчика. Ошибка, связанная с неучетом поглощения \( \beta \)-частиц в газе, будет тем меньше, чем больше энергия \( \beta \)-перехода, чем меньше исследуемого газа в камере и чем меньше ее размеры. При этом следует отметить, что само использование в качестве детектора цилиндрического \( \beta \)-счетчика, толщина стенок которого равна обычно 20—30 мкм, ограничивает применимость метода лишь случаями с достаточно большой энергией \( \beta \)-излучения. Для аргона-41 (\( E_\beta = 1,245 \)) формула была проверена экспериментально при давлении исследуемого газа в камере, равном атмосферному, при этом было получено, что погрешность не превышает 20%.

Производить абсолютные измерения газообразных активностей можно и при помощи устройства, состоящего из торцового счетчика, смотрящего внутрь цилиндрической камеры (см. рисунок 2). Расчет коэффициента \( k \)
для этого случая, при условии $R/\alpha > 5$ был произведен Михайловым и Туркиным (2). Концентрация газа здесь также принималась одинаковой по всему объему камеры *), но в отличие от первого случая, учитывалось поглощение $\beta$-частиц в заполняющем камеру газе, которое принималось подчиняющимся экспоненциальному закону. Окончательное выражение для числа частиц, проникающих через окно в счетчик, полученное в этой работе, имеет вид

\[
N(H,R) = \frac{1}{2} A \beta S \int_0^H h \, dh \left\{ \frac{e^{-\mu_0 h}}{h} - \frac{e^{-\mu_0 \sqrt{h^2 + R^2}}}{\sqrt{h^2 + R^2}} \right\} - 
\]

где:

- $A$ — активность в 1 см$^3$ газа в камере
- $S$ — площадь окошка счетчика в см$^2$
- $E_2$ — интегральная показательная функция (функция Эй)
- $\mu_0$ — коэффициент поглощения $\beta$-частиц ($\mu_0 = \mu_1 + \mu_2 \frac{t}{n}$)

*) причем рассеяние $\beta$-частиц от стенок также не учитывалось.
значения $H$, $h$ и $R$ ясны из рис. 2, $t$ — толщина слюды окна счетчика в см.
В приведенной ниже таблице собраны значения $k$ в формуле

$$A_{	ext{Кюри}} = \frac{k}{S} \frac{N_{\text{имп}}}{\text{мин}}$$
— площадь окна счетчика в см$^2$, рассчитанная по формуле (2), при
толщине слюды входного окна 5 мг/см$^2$ и при атмосферном давлении заполняющего камеру газа, представляющему собой воздух с незначительными примесями.

Следует оговорить, что данными экспериментальной проверки приведенной формулы (и таблицы) мы, к сожалению, не располагаем.

<table>
<thead>
<tr>
<th>Таблица</th>
</tr>
</thead>
<tbody>
<tr>
<td>Таблица значений $K \cdot 10^6$ для различных геометрических условий и энергий $\beta$-излучения</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$R$ (см)</th>
<th>$E$ (мэв)</th>
<th>Высота камеры $H$ см</th>
</tr>
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<td></td>
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</tr>
<tr>
<td>10</td>
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<td>3,28</td>
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<td></td>
<td>0,70</td>
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<tr>
<td></td>
<td>0,90</td>
<td>2,03</td>
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<tr>
<td></td>
<td>1,245</td>
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<td>0,35</td>
<td>2,84</td>
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<tr>
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<td>2,00</td>
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<td>0,90</td>
<td>1,69</td>
</tr>
<tr>
<td></td>
<td>1,245</td>
<td>1,60</td>
</tr>
<tr>
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<td>1,245</td>
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<td>25</td>
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<td>1,86</td>
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<tr>
<td></td>
<td>0,90</td>
<td>1,43</td>
</tr>
<tr>
<td>1,245</td>
<td>1,395</td>
<td>0,86</td>
</tr>
</tbody>
</table>

Конечно, идеальным методом являлся бы такой, когда в известном объеме считались бы все 100% происходящих в нем распадов. Эффективность близкая к 100% может быть получена при использовании счетчиков внутреннего заполнения. Ошибка в этом случае возникает за счет неучета частиц, уходящих в стенки счетчика без последующего разряда, а также за счет краевых эффектов, расчет которых крайне затруднителен. Оценка
первого из указанных эффектов для мягких β-излучателей (C^{14}, S^{35}) при стандартных заполнениях (90% Ar + 10% C_{2}H_{5}OH при p = мм Hg) и не слишком малых диаметрах счетчиков дает незначительную величину. Рецепт исключения краевых эффектов впервые был дан в работе (3). Для этого используются два соединенных между собой счетчика, все параметры которых, исключая длины катодов, одинаковы. При введении в счетчики исследуемого газа, разность скоростей счета, полученных на плато длинного и короткого счетчиков, дает число распадов в объеме, равном разности объемов обоих счетчиков (защищованная область рисунок 3). Концевые эффекты при этом исключаются. На рис. 4 схематично изображена установка, использовавшаяся авторами для проведения абсолютных измерений активности некоторых радиоактивных газов (ацетилен СО_{2}). Проба берется в кран 1. Баллон 4, порционный кран 3 и двухпроходной кран 2, дают возможность вводить в счетчики желаемую часть взятой в пробный кран активности. В запленном нижнем отростке трехпроходного крана 5 налит циклогексан, пары которого используются для заполнения счетчиков. Использование в качестве рабочего газа счетчиков чистых паров циклогексана дает ряд преимуществ. Качество плато при подобном заполнении менее чувственно к примесям посторонних газов. Система
хранения и напуска чрезвычайно проста (один трехпроходной кран). Кроме того удельная ионизация у паров циклогексана довольно большая (ее величина примерно пропорциональна числу электронов в молекуле), что снижает ошибку, связанную с уходом β-частиц в стенки счетчика без последующих

![Diagram](image_url)

Рис. № 4

![Graph](image_url)

Рис. № 5
разрядов. На рис. 5 в качестве примера приведены счетные характеристики, снятые при подобном заполнении с помощью обычной пересчетной установки.

Проверка описанного метода путем измерения удельной активности раствора Na$_2$C$^{14}$O$_3$ с проведением количественного выделения C$^{14}$O$_2$ и последующим измерением активности полученного газа на данной установке, а также с помощью газоразрядного 4π-счетчика, позволяет заключить, что суммарная ошибка определения активности данным методом не превышает 10%.

Пользуясь описанным методом двух счетчиков, авторами была определена эффективность для нескольких видов счетчиков внутреннего заполнения. Для конструкции счетчика, чертеж и параметры которого приведены на рис. 6, для газов, меченых C$^{14}$. Она больше 95%, что делает подобный счетчик вполне пригодным для проведения абсолютных измерений.

Заканчивая изложение, остановимся на абсолютных измерениях активности жидкостей, производить которые обычными методами довольно затруднительно.

В работе (4) авторами описан метод измерения активностей β-активных жидкостей, основанный на полном испарении известного количества жидкости в известный объем с последующим измерением введенной активности «газовыми» методами. При этом можно производить как относительные, так и абсолютные измерения.
На рис. 7 изображена схема установки, использовавшейся авторами для относительных измерений Cs₂⁵. Проба вводится в кран 1, который подсоединяется к установке с помощью шлифа и установка откачивается до 10⁻¹ мм Hg. Затем, при открывании крана 1 проба испаряется в объем установки. На рис. 8 дается зависимость скорости счета измерительной установки для этого случая от количества эталонного раствора Cs₂³⁵, отмеренного в пробный кран микропипеткой. Как видно, экспериментальные точки хорошо ложатся на прямую, что свидетельствует как о хорошей линейности, так и о хорошей воспроизводимости метода.

Для ряда жидкостей (этиловый, метиловый спирты, сероуглерод (C₁⁴H₂OH, C₁⁴H₃OH, Cs³⁵) подобным методом производились и абсолютные измерения и были получены хорошо воспроизводимые результаты.

Для иллюстрации остановимся на абсолютных измерениях с помощью установки, аналогичной изображенной на рис. 4, но включающей в себя две пары счетчиков, конструкция которых такая же, как и счетчика, изображенного на рис. 6.

Одна пара счетчиков имеет стальные катоды, другая — платиновые. Пробки сделаны из тefлона.

В приводимой ниже таблице даны результаты нескольких повторных измерений удельной активности одной и той же жидкости Cs₂³⁵ (b μ c/cm³).
<table>
<thead>
<tr>
<th>№№</th>
<th>Вес взятой пробы в мг: 2,2 0,9 8,2 2,8 1,7</th>
<th>Среднее значение из 5 измерений</th>
<th>Среднее отклонение</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. 2 стальных счетчика</td>
<td>77 74 78 87 73</td>
<td>77,8</td>
<td>4,7%</td>
</tr>
<tr>
<td>2. 2 платиновых счетчика</td>
<td>84 70 79 90 69</td>
<td>78,4</td>
<td>9,5%</td>
</tr>
<tr>
<td>3. 1 длинный платиновый</td>
<td>78 71 78 81 69</td>
<td>75,4</td>
<td>4,3%</td>
</tr>
<tr>
<td>4. 1 короткий платиновый</td>
<td>77 73 79 79 69</td>
<td>75,4</td>
<td>3,5%</td>
</tr>
</tbody>
</table>

В строке 1 (2 стальные счетчика) даны результаты, полученные дифференциальным методом с помощью пары счетчиков со стальными катодами.
В строке 2 (2 платиновых счетчика) — результаты, полученные дифференциальным методом с помощью пары счетчиков, имеющих платиновые катоды.
В графах 3 и 4 (1 длинный платиновый и 1 короткий платиновый) даны результаты, рассчитанные по счету длинного и короткого платиновых счетчиков, в отдельности, считая их эффективность равной 100%.
Описанные методы нашли применение в лабораторной практике, причем последние два оказались особенно полезными при измерениях активности ряда жидкских органических препаратов.

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II. 2. 12. SOME METHODS OF MEASURING $\beta$-ACTIVE GASES

V. V. BOCHKAREV, V. A. BAZHENOV

(Translated from Russian)

Carrying out absolute measurements of the activity of gaseous $\beta$ emitters involves greater difficulties than measurements of soluble solid compounds. However, various comparatively simple methods of measuring radioactive gases have recently been worked out. Some of these will be described below. They are not precision methods but the fact that they are already on hand seems to us to make them of some interest.

Absolute measurements become feasible as soon as we can determine theoretically, for a given experiment, the value of $k$ in the formula $a = kN$. i. e. the coefficient of proportionality relating the reading of the measuring instrument, $N$, to the activity of the substance being investigated, $a$.

If a thin-walled cylindrical counter of $\beta$ particles is placed in a cylindrical vessel along its axis, it forms a simple instrument for measuring the activity of the gas. A formula for determining $k$ for a given geometry was given in 1948 (1) for the following conditions and on the following assumptions: the concentration of the gas was considered constant for the whole volume enclosed by the walls of the chamber; the absorption of the $\beta$ particles in the gas in the chamber and scattering from the walls of the chamber were not taken into account; the dimensions of the counter and the chamber were governed by the equation $H - h \geq 2r$, where $r$ is the radius of the counter, $H$ is the half-height of the chamber and $h$ is half the length of the working volume of the counter (see figure 1). This resulted in a fairly simple equation, relating the number of particles hitting the lateral surface of the counter $N_0$ to the number of disintegrations per second in 1 cm$^3$ of the volume of the chamber $n_0$, the accuracy increasing as the ratio between the radius of the chamber and the radius of the counter $R/r$, rose. When $R/r = 5$ the margin of error does not exceed 6.7%.

$$N_0 = \frac{n_0 r}{2} \left\{ \sqrt{R^2 - r^2} \left( \sqrt{R^2 - r^2 + (H + h)^2} - \sqrt{R^2 - r^2 + (H - h)^2} \right) + 
+ (H + h)^2 \ln \frac{\sqrt{R^2 - r^2 + (H + h)^2} + \sqrt{R^2 - r^2}}{H + h} 
- (H - h)^2 \ln \frac{\sqrt{R^2 - r^2 + (H - h)^2}}{H - h} 
- (R - r) \left( \sqrt{(R - r)^2 + (H + h)^2} - \sqrt{(R - r)^2 + (H - h)^2} \right) + 
+ (H + h)^2 \ln \frac{\sqrt{(R - r)^2 + (H + h)^2} + (R - r)}{H + h} 
- (H - h)^2 \ln \frac{\sqrt{(R - r)^2 + (H - h)^2} + (R - r)}{H - h} \right\}; \quad (1)$$

In order to calculate the number of particles registered by the counter per second, it is necessary to make a correction by the usual methods for absorption.
in the walls of the counter. The size of the error caused by failure to allow for absorption of \( \beta \) particles in the gas will be inversely proportional to the energy of the \( \beta \) transfer and proportional to the pressure of the gas under investigation in the chamber and to the dimensions of the chamber. It should be noted, in this connexion that the use as detector of a cylindrical \( \beta \) counter, the wall-thickness of which is usually 2—30 mg/cm\(^2\), means that this method can be used only in cases where there is a sufficiently high \( \beta \) emission energy. Using \( \text{A}^{41} \) (\( E_\beta = 1.245 \)) the equation was checked experimentally, with the gas under investigation in the chamber at atmospheric pressure, and it was found that the error did not exceed 20%.

Absolute measurements of gaseous activity are also possible with the help of a unit consisting of an end-window counter recording inside a cylindrical chamber (see figure 2). MIKHAILOV and TURKIN \(^{2}\) worked out a value for the coefficient \( k \) in regard to this method, taking \( R \leq 5 \). In this case also, the gas concentration was considered as constant over the whole volume of the chamber \(^{\ast}\) but, unlike in the previous case, account was taken of \( \beta \) particle absorption in the gas filling the chamber; this absorption was taken as obeying an exponential law. The formula finally arrived at in this paper for the number of particles penetrating the counter window is

\[
N (H,R) = \frac{1}{2} A_\beta S \int_0^H h \, dh \left\{ e^{-\mu_0 \frac{h}{h}} - e^{-\mu_0 \sqrt{h^2 + R^2}} \right\} - \mu_0 \left[ -E_2 (\mu_0 h) + E_2 \left( -\mu_0 \sqrt{h^2 + R^2} \right) \right]
\]

where:

\[ A_\beta = \text{activity in 1 cm}^3 \text{ of the gas in the chamber} \]
\[ S = \text{area of the counter window in cm}^2 \]
\[ E_i = \text{integral exponential function} \]
\[ \mu_0 = \text{coefficient of } \beta \text{ particle absorption} \]

The meaning of \( H \), \( h \) and \( R \) is clear from figure 2; \( t = \text{thickness of the mica window of the counter in cm} \).

In the table set out below are given values for \( k \) in the equation:

\[
A \text{ curies} \frac{\text{litres}}{\text{minutes}} = k \frac{N \text{ impulses}}{S \text{ cm}^2}
\]

where \( S \) represents the area of the counter window in cm\(^2\). These values are calculated according to formula (2), taking the thickness of mica entry-window as 5 mg/cm\(^2\) and the pressure of the gas in the chamber as equal to that of air containing insignificant impurities.

Unfortunately we do not have the data providing experimental proof of the formula and table given.

\(^{\ast}\) The scattering of \( \beta \) particles from the walls was also ignored.
### Table of $k \cdot 10^{10}$ values for various geometrical conditions and β-radiation energies

<table>
<thead>
<tr>
<th>R (in cm)</th>
<th>E (in MeV)</th>
<th>Height of the chamber H (in cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>10</td>
</tr>
<tr>
<td>10</td>
<td>0.35</td>
<td>3.28</td>
</tr>
<tr>
<td></td>
<td>0.70</td>
<td>2.40</td>
</tr>
<tr>
<td></td>
<td>0.90</td>
<td>2.03</td>
</tr>
<tr>
<td></td>
<td>1.245</td>
<td>1.94</td>
</tr>
<tr>
<td>15</td>
<td>0.35</td>
<td>2.84</td>
</tr>
<tr>
<td></td>
<td>0.70</td>
<td>2.00</td>
</tr>
<tr>
<td></td>
<td>0.90</td>
<td>1.69</td>
</tr>
<tr>
<td></td>
<td>1.245</td>
<td>1.60</td>
</tr>
<tr>
<td>20</td>
<td>0.35</td>
<td>2.76</td>
</tr>
<tr>
<td></td>
<td>0.70</td>
<td>1.89</td>
</tr>
<tr>
<td></td>
<td>0.90</td>
<td>1.57</td>
</tr>
<tr>
<td></td>
<td>1.245</td>
<td>1.51</td>
</tr>
<tr>
<td>25</td>
<td>0.35</td>
<td>1.86</td>
</tr>
<tr>
<td></td>
<td>0.70</td>
<td>1.50</td>
</tr>
<tr>
<td></td>
<td>0.90</td>
<td>1.43</td>
</tr>
<tr>
<td></td>
<td>1.245</td>
<td>1.43</td>
</tr>
<tr>
<td>30</td>
<td>0.35</td>
<td>1.61</td>
</tr>
<tr>
<td></td>
<td>0.70</td>
<td>1.47</td>
</tr>
<tr>
<td></td>
<td>0.90</td>
<td>1.47</td>
</tr>
<tr>
<td></td>
<td>1.245</td>
<td>1.395</td>
</tr>
</tbody>
</table>

An ideal method would, of course, be one in which 100% of the disintegrations occurring in a given volume were counted. An efficiency of nearly 100% can be achieved using counters with an inner filling. Errors then arise from uncounted particles escaping into the walls of the counter without a corresponding discharge, and also from the end-effects, the calculation of which is extremely difficult. Calculation of the first of the above-mentioned effects for soft β emitters ($\text{C}^{14}$, $\text{S}^{35}$) with standard fillings (90% Ar + 10% C$_2$H$_5$OH at 100 mm Hg pressure) and counters of not too small diameter gives insignificant values. A method of excluding end-effects was first published in paper (3). Two linked counters were used, identical except as regards the length of the cathodes. On the gas being introduced into the counters, the difference in the count speed obtained on the plateau of the long and of the short counter gives the figure for the disintegrations in a volume equal to the difference in volume of the two counters (hatched area in figure 3). End-effects are thus eliminated.

Figure 4 shows schematically the apparatus which was used by us for making absolute measurements of the activity of certain radioactive gases (acetylene-CO$_2$). The sample is introduced at cock 1. The flask 4, the batch cock 3 and the 2-way cock 2 make it possible to pass into the counters any desired quantity of the active material introduced into cock 1. Cyclohexane flows in the sealed lower outlet of the 3-way cock 5. Its vapour is used for filling the counters. To use pure vapourized cyclohexane as the working gas in the counters gives many advantages. The quality of the plateau obtained by using such fillings is less sensitive to an admixture of extraneous gases. The system of storage and fillings is extremely simple (one 3-way cock). Furthermore, the
specific ionization of the cyclohexane vapour is fairly great (its extent being approximately proportional to the number of electrons in the molecule); this reduces the error resulting from the escape of \( \beta \) particles into the walls of the counter without corresponding discharges. In figure 5 are given, as an example, data regarding the plateaux obtained for such a filling by means of the normal type of scaler-unit.

The method described can be verified by measuring the specific activity of a solution of \( \text{Na}_2\text{C}^{14}\text{O}_3 \), including the amount of \( \text{C}^{14}\text{O}_2 \) evolved, and then measuring the activity of the gas obtained by means of the apparatus described, and also by means of a gaseous 4 \( \pi \) counter. The results obtained indicate that the total error entailed in determining activity by this method does not exceed 10%.

Using the two-counter method described above, we determined the efficiency of certain types of filled counters. For a counter designed to the specifications given in figure 6, filled with gases labelled with \( \text{C}^{14} \), the efficiency is more than 95\%, which makes this type of counter perfectly suitable for carrying out absolute measurements.

In conclusion let us say a few words on absolute measurements of the activity of liquids — an operation which presents considerable difficulties using the normal methods.

In another paper \(^4\) we have described a method of measuring the activity of \( \beta \) active liquids based on the complete evaporation of a known quantity of liquid into a known volume with subsequent measurement of the induced activity by "gaseous" methods. It is possible to carry out both comparative and absolute measurements.

Figure 7 shows the lay-out of the apparatus we used for the comparative measurement of \( \text{CS}^{35}_2 \). The sample is introduced at cock 1, which is connected to the apparatus by a ground joint, and the apparatus is evacuated up to \( 10^{-1} \) mm Hg. When cock 1 is opened, the sample is vaporized into the volume of the apparatus. Figure 8 shows how the counting rate of the measuring apparatus depends in this case on the amount of calibrating solution \( \text{CS}^{35}_2 \), measured off into the test cock by a micropipette. It will be seen that the experimental readings lie close to a straight line which points to the good linearity and the good reproducibility of the method.

A similar method was also used to take absolute measurements for various liquids — ethyl, methyl alcohol, carbon bisulfide (\( \text{C}_4\text{H}_8\text{OH}, \text{C}^{14}\text{H}_8\text{OH}, \text{CS}^{14}_2 \)) — and gave highly reproducible results.

Let us take as an illustration the absolute measurements obtained with an apparatus similar to that shown in figure 4, but including two pairs of counters, the design of which was the same as that of the single counter shown in figure 6.

One pair of counters had steel cathodes, the other platinum. The stoppers were made of teflon.

In the table given below are shown the results of certain repeated measurements of the specific activity of a single liquid \( \text{CS}^{35}_2 \) (in \( \mu\text{c/cm}^2 \)).
<table>
<thead>
<tr>
<th>Test No.</th>
<th>Weight of the sample in mg:</th>
<th>2.2</th>
<th>0.9</th>
<th>8.2</th>
<th>2.8</th>
<th>1.7</th>
<th>Average figure for five measurements</th>
<th>Average deviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Two &quot;steel&quot; counters</td>
<td>77</td>
<td>74</td>
<td>78</td>
<td>87</td>
<td>73</td>
<td>77.8</td>
<td>4.7%</td>
</tr>
<tr>
<td>2</td>
<td>Two &quot;platinum&quot; counters</td>
<td>84</td>
<td>70</td>
<td>79</td>
<td>90</td>
<td>69</td>
<td>78.4</td>
<td>9.5%</td>
</tr>
<tr>
<td>3</td>
<td>One long &quot;platinum&quot; counter</td>
<td>78</td>
<td>71</td>
<td>78</td>
<td>81</td>
<td>69</td>
<td>75.4</td>
<td>4.3%</td>
</tr>
<tr>
<td>4</td>
<td>One short &quot;platinum&quot; counter</td>
<td>77</td>
<td>73</td>
<td>79</td>
<td>79</td>
<td>69</td>
<td>75.4</td>
<td>3.5%</td>
</tr>
</tbody>
</table>

For test 1 (2 "steel" counters) are given the results obtained by the differential method using a pair of counters with steel cathodes.

For test 2 (2 "platinum" counters) are given the results obtained by the differential method using a pair of counters with platinum cathodes.

For tests 3 and 4 (1 long "platinum" and 1 short "platinum" counter) are given the results obtained by using, first, one long and, then, one short platinum counter, taking the efficiency of the counters as 100%.

The methods described have been used in laboratory practice and the last two have proved particularly useful in measuring the activity of a number of organic liquids.

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II. 2. 13. ON THE SINGLE-SCINTILLATOR COINCIDENCE TECHNIQUE

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INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, AUSTRIA

Abstract

It is shown that in low-energy counting with single scintillators the coincidence technique can be improved by summing the pulses from the photomultipliers in coincidence. The use of semiconductor devices in the circuitry is advocated for simplicity and reliability. Further development in this technique concerns the scintillator and the photocathode.

Sur la technique de coincidence à l'aide d’un scintillateur unique.

L’auteur montre que dans le comptage de particules de basse énergie, à l’aide d’un scintillateur unique, il est possible d’améliorer la technique des incidences en additionnant les impulsions des deux photomultiplicateurs en coincidence. L’utilisation de circuits électroniques à semi-conducteurs est recommandée pour sa simplicité et sa sûreté de fonctionnement. D’autres améliorations de cette technique ont trait aux scintillateurs et à la photocathode.

Техника использования одного сцинтилятора на принципе совпадения

Показано, что технику счета с малым энергетическим излучением одного сцинтиллятором можно улучшить с помощью суммирования импульсов из фотоумножителей совпадения. Для простоты и надежности электронной схемы рекомендуется использование полупроводников. Дальнейшее усовершенствование этой техники касается сцинтиллятора и фотокатода.

Sobre la técnica de coincidencias mediante monocentelleadores

El autor demuestra que en el recuento de partículas de baja energía mediante monocentelleadores es posible mejorar la técnica de coincidencias sumando los impulso procedentes de los fotomultiplicadores conectados en coincidencia. Por su sencillez y funcionamiento seguro, recomienda el empleo de semiconductores en los circuitos electrónicos. Otros perfeccionamientos de esta técnica se refieren al centelleador y al fotocátodo.

The coincidence technique has become an established tool in low-energy scintillation counting. The straight-forward, single-photomultiplier technique, however, is still preferred by some, presumably on account of the larger pulses obtained due to the more efficient light collection and the simplification in the counting electronics. This is particularly true if an efficient scintillator is combined with a low-noise photomultiplier, well-cooled, and sufficient activity is on hand.

However, these are no valid reasons to reject the coincidence technique and its obvious advantages. By summing the two pulses from the photomultipliers in coincidence one actually utilizes light collection in both directions. On the other hand, the application of transistors in coincidence circuits
simplifies the electronics and increases the reliability of the technique. Both innovations result in an improved figure of merit \(S^2/B\) as the counting rate \(S\) of the signal is increased at the same time as the background \(B\) in considerably diminished.

In effect, this means that one can lower the discrimination level at which the energy spectrum measured is cut off. This is important in C\(^{14}\) liquid-scintillation counting where the spectrum at the noise threshold of present photomultipliers still increases in intensity. In tritium low-level counting, on the other hand, this is essential as the \(\beta\) spectrum is masked to a considerable extent by the photomultiplier noise. A more complete count of \(\beta\) particles is thus achieved, the cut-off energy level being more than halved. Thus the conditions for low-level counting according to the criterion \(4\sigma = B\) are improved.

In figure 1 a block scheme of the arrangement is shown. This is a single-scintillator variation of a two-crystal system used in an XYZ-pulse-height analyser by the author where the summing of the pulses is similar to that employed in the sum-coincidence technique \(^{(1)}\). In the single-scintillator case it is not necessary to compensate for differences in scintillator properties, as the same scintillator feeds both photomultipliers. The latter two should, however, possess comparable amplification factors. An additional feature is the possibility one has of checking one photomultiplier against the other for drift, thus increasing assurance of reliability.

The pulses from the White cathode-followers are summed either in an ohmic circuit or with the help of diodes. To the output can be connected a cathode-ray oscilloscope providing it is sensitive and fast enough, otherwise it must be placed after a linear amplifier. The amplification of the photo-

![Figure 1. Block scheme of single-crystal coincidence technique.](image-url)
multipliers can be equalized by adjusting the high tension of one of the photomultipliers so that pulses of the same energy when coming through the two channels are equal. If a suitable isotope is not contained in the scintillator a γ source can be placed outside or use can be made of the noise pulses.

To the equalizers of the anode pulses of the two photomultipliers a fast coincidence unit is connected, the output pulse of which can be used to operate the gate of the counting equipment. In low-energy β counting liquid organic scintillators are used, in which case the pulse lengths are usually of the same order as the time resolution of fast coincidence circuits. It is therefore advisable to use fast coincidence and anti-coincidence units and gates (2) so as not to shorten unnecessarily the live time of counting. This is also true with respect to the differential discriminator utilized to sample the desired part of the spectrum from the rest of the pulses. This holds even in low-level counting if the high-energy background is of concern.

At present it is possible to use transistors in such fast circuits advantageously (3) (4). In the last-named reference a fast coincidence circuit is described which could be adopted for the above purposes. The input resolving time of the unit is some nanoseconds and its discrimination level can be adjusted to tens of millivolts, if needed, while the output pulse is some volts. Two such units could be combined to form a fast differential discriminator.

Finally some remarks should be made on the efficiency of low-energy counting with the single-scintillator coincidence technique. Because the number of quanta liberated is low and the efficiency of photocathodes is not one hundred percent, there exists an optimum number of coincidence channels that should be used. This number decreases with diminishing number of quanta and decreasing efficiency of the photocathodes, and thus if there was no noise the single photomultiplier system would be the only reasonable one (see first paragraph). On the other hand, the degree of noise suppression (in direct relation to the number of chance coincidences) is higher the greater the number of coincidence channels. Hence, as we are concerned with the lowest radiation energies measurable with scintillators in the region of noise, it is reasonable to conclude that the two-channel coincidence technique is the most efficient single-scintillator counting arrangement, when the number of noise pulses is not prohibitive. Thus as a practical minimum we must have at least two quanta which release a minimum of one electron per photocathode respectively in order to register a particle. This presupposes that the predominant noise is due to single thermal electrons from the photocathode.

We may thus summarize that the application of a two-channel coincidence technique together with fast discrimination of sum-pulses from the two channels is the optimum. Further development in low-energy scintillation counting should therefore be in the direction of improving the sensitivity and reducing the noise generation of the photocathodes, in addition to any improvement in the efficiency of the scintillator and the light collection from it.

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DISCUSSION

The Chairman (Mr. J. Domanus, Poland):

Now we will go on to the discussion of all the papers presented today. The first question is from Dr. Putman to Dr. Pate.

Dr. Putman (United Kingdom):

In correcting for absorption by the source support in 4π counters, we found that linear extrapolation of the absorption curve is not justified below about 2% of the absorption half-thickness. In such thin films, the loss of counts on one side is largely compensated by back-scattering to the other side and experimental curves with \( P^{32} \) level off for low thickness. The error involved in extrapolating linearly was in some cases of the order of 1%. Did Dr. Pate or Dr. Campion investigate this effect?

Dr. Pate (United States of America):

As the author of the review paper I should perhaps quote general experience rather than just my own. Dr. Putman and I put our heads together and figured out what the superficial density of the source mount would need to be to correspond to this 2% of the half-thickness in the case of S\(^{35}\), and on the basis of some absorption coefficient figures he produced from his memory, it comes out as something like 40 microgrammes per square centimetre. Now there have been certainly more than one series of experiments in which the counting rate of a 4π counter was measured as a function of source-mount thickness which went down to film thicknesses well below this limit, and none of these showed the effect to which he refers, as far as I am aware, in the case of low-energy β emitters. In the case of higher-energy β emitters, of course, the lowest thickness of film used was an even smaller fraction of the half-thickness. The extrapolation to which he refers is, of course, not linear in the general case; it is curved. I presume he means that the curvature is in a reserve sense to that in the curves I showed. I am not aware of any experience of the type he mentions, possibly Dr. Campion may be.

Dr. Campion (Canada):

No, I have not observed anything like a flattening-out of the function of counting rate against source thickness.

Dr. Putman:

Dr. Pate repeatedly referred to the preparation of “carrier-free” sources for 4π counters. How does he guard against loss of activity by absorption or adsorption to the walls of containers? D. B. Smith found that serious losses occur with solutions containing a few parts per million of carrier, although the losses may be reduced by coating glass containers with silicone enamel. Were the samples which you exchanged with other users in the form of deposited sources or solutions and were different or identical methods of measurement used?

Can Dr. Pate say what is the effect of using carrier-free solutions on the preparation of standards or sources for comparison?

Dr. Pate:

This question is obviously rhetorical. Dr. Putman knows perfectly well what the answer is, and I think we all do. Let me make two comments. Carrier-free sources, once they are laid down on the source mount, can just as easily be counted. One can make counting rate measurements on them just

336
as well, once they are on the film, as in the case of sources with carrier present in them. One says nothing about the meaning of such a counting rate, however. I think Dr. Putman misunderstood my remarks in the paper when I referred to the situation that would exist, for example, in various source-preparation methods in the absence of carrier. I was merely illustrating the limit when one reduces the amount of carrier present in a particular determination. I was not advocating the circulation of solution standards in the absence of carriers. This is well known to be dangerous. I would like to add something to my reply to the first question. Dr. Putman made mention of a study he has made of the back-scattering process in $4\pi$ counters which apparently gives him additional evidence of this flattening of the source-mount absorption function. He made mention of the fact that he thought this flattening that he had observed was due to a compensation, the effect of back-scattering from the film reducing the absorption of $\beta$ particles in thin source mounts. There have, of course, been many such studies made. The names of Sousa and Charback come to mind. There was some contribution to this from McGill. The works to which I referred were also done with very thin source mounts which were in fact a very small fraction, a smaller fraction than 2% of the half-thickness, and indeed no such effect was observed. In the case of Sousa and Charback I believe they were using Geiger-Müller counters. The work at McGill was done using proportional counters. In no case was any such effect observed.

Mr. Curran (United Kingdom):

Mr. Pate stressed the advisability in $4\pi$ counting of having the field strength high in the vicinity of the source. This is clearly advantageous, yet in the $4\pi$ counter shown by him we have two thin wire loops and within these rather large loops there must exist a node of field strength — a region of close to zero field. This seems to me very questionable practice in absolute work of high accuracy. Has any measurement of the field configuration been made? Is it not better to stay with systems so measured if none have been made for loop counters? Personally I feel this is a situation which leads me to favour co-axial or approximately co-axial fields.

Dr. Pate:

Let me first make one comment. It seems to me that the questions which are being addressed to me as the author of this review paper ought properly to be asked of this whole group present. I will attempt to answer these questions, but it is entirely possible that the other people present will have better answers than I, since I do not in the few seconds available have access to a library to look up answers that I do not know. The only work of which I am aware on field strengths in counters employing ring-shaped anodes was done in connexion with the work in the laboratory of Dr. Wiedenberg and his students. This was not particularly applicable to the present case because the anode ring in that case lay in a plane parallel to that of the source, whereas the present situation that we are discussing is one in which the anode ring lies at right-angles to this plane. As far as I am aware, and the majority of people will possibly confirm this, the evidence which one must produce as to the efficacy or otherwise of such a counter system is entirely experimental. The results with this type of counter with a ring anode are found to be in excellent agreement with the types of counter which the questioner would perhaps prefer.
Dr. Yaffe (Canada):

I might perhaps add a little to that, if Dr. Pate does not mind. We have in our laboratory at McGill University both the loop counter, of which you have seen several pictures, and the pill-box type of counter which Dr. Curran prefers, and we have taken the same sample and measured this in each counter with identical results within the statistical margin of error. We have not, however, investigated the field strength within the counter itself.

Dr. Curran:

I would just like to make one remark before we close the discussion on this particular question. My question was concerned wholly with the very high accuracy now being asked of the loop counter — this "better than 1%" question. I am quite satisfied that both types of counter to which we have been referring are good to about 1%, but I am not sure that any of us are clear as to the variations at better than 1%. I thought it was about time that further work could usefully be done on these field questions.

Dr. Pate:

Dr. Yaffe will possibly confirm my recollection about the agreement between counting-rate measurements between the ring-type counter and, for example, a completely different disintegration-rate determination method. I refer to a measurement on Au\(^{198}\), some considerable time ago, which would lead one to suppose that the agreement would be much better than 1%. More direct data on this question would probably be provided also by some experience at the Brookhaven National Laboratory, where we also had both ring-type-anode counters and pill-box counters. We have also counted identical sources in both types of counter and found agreement to be, within the statistical error, at about 0.1 or 0.2%.

Mr. Braun (Sweden):

What is the accuracy with which you can measure, by the methods given in your paper, the amount of active material evaporated onto your films?

Dr. Yaffe:

This is a question for an analytical chemist. Standards of various solutions are established, the spectrophotometric analysis of the transmission is carried out at a particular wave-length, and the reproducibility is determined. I do not have any figures with me at the moment, but I would be quite happy to give Mr. Braun some later. Spectrophotometric analysis is a standard technique that can be done very accurately — certainly to less than a fraction of 1% — if pains are taken with the standard solutions, and it will vary with the material which is being used.

The Chairman:

Does that answer your question?

Mr. Braun:

Partly.

Dr. Curran:

I was very much inclined to think, on hearing Dr. Manov discuss the problem of pure materials — i.e. the difficulty of obtaining the very many materials which go into making an accurate measurement from a source — materials for the source itself, the support and the many components of the counter — that his statement of the problems made a very good case indeed for the use of an effectively "wall-less" counter. It is also, to my mind, a
fairly good argument in favour of doing work of an absolute nature with material in the gaseous phase, the gas being isolated as far as possible from walls or any other material. This is partly a comment and partly to ask Dr. Manov whether he would agree that the problem has become so serious as to justify an attack along the lines of "wall-less" counting.

Dr. Manov (United States of America):

I quite agree with Dr. Curran that if we could have a "wall-less" counter it would be an extremely good one. At the same time we must have some kind of anti-coincidence shielding if we are to work in the very low-level region of from zero to approximately ten disintegrations per minute, and one must therefore have some way of keeping the γ energy, for example, from the innermost shield from getting into the "wall-less" counter. The best thing I can think of is to have the innermost shield made of mercury enclosed in a plastic container made of old carbon. If we could get a "wall-less" counter for all our isotope measurements, it would indeed be a step forward.

Dr. Mann (United States of America):

Could Dr. Manov please describe the geometry pertaining to the results of counts per minute per gramme given in Table III of his paper?

Dr. Manov:

I would be happy to. We measured these. These are counts per minute above background, using a 3" × 3" crystal and a Dumont photomultiplier tube, the whole arrangement being inside 15 cm of iron and about 3 cm of mercury.

Dr. Mann:

You mention blocks of steel and blocks of lead; I was wondering whether this is a surface count or a count completely due to the solution determining the total disintegrations per gramme.

Dr. Manov:

I am sorry, I misunderstood your question.

Taking as an example item No. 1 in Table III — electrolytic copper foil 1 cpm per gramme — this would be on a sheet of about 0.0005" thick in the appropriate area. Other items, like adhesive cellulose tape, are given in sq. cm. of area, etc.

Dr. Mann:

A question to Mr. Robinson, please. What is the α count rate and also the total number of events counted in a typical observation?

Dr. H. P. Robinson (United States of America):

The α count rate on all these points shown on the curves was approximately 40 000 cpm. The total number of counts taken per point varied from $2 \times 10^6$ to approximately $12 \times 10^6$ for an average value of perhaps $5 \times 10^6$ per point.

Professor Pappas (Norway):

If I understood Mr. Steyn right, he measures the uranium content chemically by igniting the precipitate and weighing it as UsOs. It is, however, difficult to obtain the right stoichiometrical composition to better than about 1% by this means unless special precautions are taken. My question is whether the stoichiometrical composition of the UsOs obtained has been checked.

Dr. Steyn (Union of South Africa):

The answer to that is that this part of the work was not done by myself,
but I had the fullest confidence in the methods used by my colleague, Mr. Strelow, who is not here today, and in the results he gave me. I am afraid I will be unable to give you any more information about that point of U$_3$O$_5$ immediately, but when I get back to South Africa I could clear this point up and maybe write to you, if you will agree to that.

Dr. Baptista (Portugal):

Certainly Dr. Yaffe’s contribution is an important step towards solving the problem of self-absorption in 4π counting methods.

Dr. Yaffe said that the evaporation technique he used for the preparation of sources can be made quantitative. Is it to be understood that he can deposit any desired amount of material or that he can transfer all the material he puts into the crucible without losses?

Since it is sometimes necessary to calibrate a given solution, does he find that the chemical manipulations necessary to obtain the radionuclide in a form suitable for evaporation can also result in a 100% transfer of the radionuclide to be measured at the source?

Dr. Yaffe:

The answer to the first part is that one places a certain amount in the crucible and this is completely transferred to the film. Now I’m not certain that I understood the second portion. Do you mean that there are losses in transferring to the crucible?

Dr. Baptista:

No; suppose I gave you a cobalt fluoride solution to calibrate and that, of course, I wanted to use your data and check the particular absorption on the curve, I then have to transform these cobalt fluorides into a chemical form suitable to form the source; that is the only way I have to apply your results. Do you think that this chemical manipulation can be done with an error less than the one we tried to correct?

Dr. Yaffe:

Well, here in effect of course all you have to do is to compare the two counting rates, before and after, using the Co$^{60}$ itself as a tracer, and the error here is certainly less than the absolute rate.

Dr. Baptista:

Well, I had thought that the numbers must be different because the source conditions are different in the first case from those in the second case and I do not know what the correction is that I have to apply, because I can make an error in the transfer.

Dr. Yaffe:

One obtains a disintegration rate per unit weight of material recovered and one can then, by a separate measurement, determine how much cobalt fluoride one had initially and how much of it has been transformed to your organic compound. Maybe I am being obtuse here, I do not see the difficulty. Do you have anything to add here, Mr. Pate?

Dr. Pate:

I have two comments: firstly, the type of compound that I think Dr. Yaffe has been working on can frequently be prepared directly in the distillation crucible; this, for example, was the case with the compound used for Ni$^{63}$, which was nickel dimethyl dioxene. This was prepared from an aliquot of a stock solution of Ni$^{63}$ as chloride which was micro-pipetted
into the distillation crucible, the dimethyl dioxene compound was prepared by adding suitable reagents, then the nickel dimethyl dioxene was distilled out of the crucible, since it was the most volatile material there.

In the general case, however, surely it is possible, in the case of a solution containing carrier, to determine the concentration of carrier on an aliquot of the solution beforehand, then determine the disintegration rate as a function of unit carrier weight, then you have the information that Dr. Baptista wants, I think.

**Dr. Baptista:**

If I sent Mr. Pate an aqueous sample of cobalt chloride and asked what is the specific activity of this sample, Mr. Pate could not use this direct solution to make the measurement if he tried to correct for self-absorption, but would have to transform this solution into another one. The problem I have is whether, if in this transformation you can be sure not to lose any material, you can be sure that in relating the final result to the primary solution you really are measuring the primary solution.

**Dr. Pate:**

Will Dr. Baptista tell me whether this hypothetical solution is large enough to allow me to take an aliquot beforehand and determine the inactive cobalt content?

**Dr. Baptista:**

Yes.

**Dr. Pate:**

Then I think there is no difficulty. Maybe I too am being obtuse.

**Dr. Yaffe:**

I think this is a problem which perennially arises between chemists and physicists, Mr. Chairman, and I think perhaps we might straighten this out and convince Dr. Baptista that this is indeed a possibility after the conference.

**The Chairman:**

Thank you very much. I do not think we will go any further in this discussion and maybe those three gentlemen would meet after the meeting and solve these problems among themselves.

**Dr. Campion:**

Since the United States National Bureau of Standards' internal gas counters are filled by placing them in a common enclosure, then, in the case of nuclides which have a $\gamma$-ray in their decay scheme, one should take into account the $\gamma$-ray sensitivity of the gas counters. This is probably a small effect for Kr$^{85}$, since the $\gamma$-ray occurs only in a small fraction of the decays, but it would be important for, say, Ar$^{41}$. However, it would be interesting to know if this has been considered.

**Dr. Mann:**

Yes, Mr. Chairman; in fact, we went one stage further: we considered the bremsstrahlung from Kr$^{85}$ and carried on experiments in which we set up a reproduction of the three counters and then filled one of the counters with methane, allowed methane to flow through it and used this as a counter, and then put the specimen of Kr$^{85}$ all around the counter to see the effect. That is way below the statistical accuracy of our experiments; I imagine that the sensitivity to $\gamma$-rays is something like a thousandth of that, at least in the case of the $\beta$ particle counters, which are, we assume,
100% efficient to β particles, and also the effect of bremsstrahlung was completely negligible.

Dr. Grinberg (France) (Translated from French):

Why is there zero back-scattering for π geometry? I understood Dr. H. P. Robinson to say this morning that the back-scattering in a π angle was equal to zero. I did not clearly understand the reasons, however. Does that depend on experimental conditions since, a short time before that, Dr. Robinson had told us that the general back-scattering was of the order of 3—4%, but that, with a π angle, it was zero.

Dr. H. P. Robinson:

The 1 π geometry counter has an acceptance angle of 120°. This means that no particles are accepted into the sensitive area that are emitted at an angle lower than 30°. It is found experimentally that essentially all back-scattered α particles are emitted at angles below 30°; therefore a 1 π counter will never see any back-scattered particles.

Dr. Reynolds (United States of America):

Dr. Mann mentioned the presence of an impurity in the C¹⁴ supplied to him. Unfortunately, I did not understand the remark and I wonder if he could clarify it.

Dr. Mann:

Actually the remark was not in a very serious vein. I was referring to the previous day when both you and Mr. Putman had been asked to confirm how the radiochemical impurities in our standards were checked. I have obtained a value for the half-life of C¹⁴ which was low by an amount which I am embarrassed to state. This could be explained by the presence of a β emitter in the sample which we obtained and, as I say, the remark about your perhaps being able to give me a statement on radiochemical purity was in a not very serious vein.
II. 3. ELECTRON-CAPTURE NUCLIDES
(SESSION 5, 16 OCTOBER 1959)

II. 3. 1. SOME REMARKS ON THE PROGRESS IN ELECTRON CAPTURE STANDARDIZATION

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Abstract

A brief review is given of the measurement of low-Z, electron-capture nuclides using the $4\pi$ X-$\gamma$ coincidence method and a pressurized (up to 4 atmospheres) $4\pi$ proportional counter. A new steel $4\pi$ proportional counter is described. This counter is designed to operate at pressures up to approximately 40 atmospheres. Results on low-Z nuclides in this counter and in the lower pressure model are compared.

The measurement of nuclides of higher Z is described. Samples have been investigated using the high-pressure counter for X-ray determination and by $4\pi$ scintillation counting employing both NaI phosphors and plastic scintillators. An attempt has been made to calibrate the samples using a liquid scintillation counter. A determination of the relative numbers of K and L X-rays emitted is described together with a measurement of the number of K X-rays accompanied by L X-rays. Accurate determination of these quantities enables high-Z, electron-capture nuclides to be assayed by the relatively simple method of $4\pi$ counting its L X-rays. The measurement of Auger electrons from the high-Z nuclides in a conventional (low pressure) $4\pi$ GM counter is also included.

A short account of a redetermination of the electron-capture/$\beta^+$ branching ratio in Co$^{58}$ is described. The method involves absolute $\beta^+$ counting and coincidence techniques.

Progrès accomplis dans l'étalonnage des nuclides à capture électronique

L'auteur donne un aperçu de la mesure de nuclides à capture électronique de nombre atomique peu élevé, par la méthode des coïncidences $4\pi$ X-$\gamma$, au moyen d'un compteur $4\pi$ proportionnel sous pression (jusqu'à 4 atmosphères). Il décrit un nouveau compteur $4\pi$ proportionnel en acier, destiné à fonctionner
sous des pressions pouvant atteindre 40 atmosphères environ. Il compare les résultats obtenus, pour des nuclides de nombre atomique peu élevé, au moyen de ce compteur et du modèle à pression plus basse.

L’auteur décrit ensuite la mesure de nuclides de nombre atomique plus élevé. Des échantillons ont été étudiés au moyen du compteur à haute pression, pour la détermination des rayons X, ainsi que par comptage $4\pi$ à scintillations, en utilisant aussi bien des cristaux de NaI que des matières plastiques. On a cherché à calibrer les échantillons au moyen d’un compteur à scintillateur liquide. L’auteur décrit ensuite comment il a déterminé le rapport numérique entre les rayons X-K et X-L émis, et comment il a mesuré le nombre de rayons X-K accompagnés de rayons X-L. Une détermination précise de ces quantités permet d’étalonner les nuclides à capture électronique de nombre atomique élevé, par la méthode relativement simple du comptage $4\pi$ des rayons X-L. Le mémoire contient aussi la description de la mesure d’électrons d’Auger émis par les nuclides de nombre atomique élevé, au moyen d’un compteur GM $4\pi$ classique (à basse pression).

L’auteur expose brièvement comment il a pu déterminer à nouveau, pour le Co$^{58}$, le rapport de bifurcation capture électronique/β+. Cette méthode comporte un comptage β+ absolu et l’emploi de techniques de coincidences.

Некоторые замечания относительно прогресса в области стандартизации захвата электронов

Приводится краткое описание измерения захвата электронов у изотопов с низким Z с помощью метода $4\pi$ X-γ-счета на совпадениях и $4\pi$ пропорционального счетчика под давлением (до 4 атмосфер). Дается описание нового стального $4\pi$ пропорционального счетчика. Этот счетчик предназначен для работы при давлении примерно до 40 атмосфер. Сравниваются результаты для изотопов с низким Z в этом счетчике и в модели низкого давления.

Дается описание измерения для изотопов с высоким Z. Были исследованы образцы с использованием счетчика высокого давления для исследования рентгеновских лучей и с помощью $4\pi$ сцинтилляционного счетчика, применяющего как фосфористое вещество NaI, так и пластические сцинтилляторы. Была предпринята попытка калибровать образцы с помощью жидкого сцинтилляционного счетчика. Определение относительного числа испускаемых K- и L-рентгеновских лучей описано вместе с измерением числа K-рентгеновских лучей, которые сопровождаются L-рентгеновскими лучами. Точное определение этих количеств дает возможность испытать изотопы с электронным захватом и высоким Z сравнительно простым методом $4\pi$ подсчета его L-рентгеновских лучей. Включено также измерение Оже-электронов из изотопов с высоким Z в обычном $4\pi$ счетчике Гейгера-Мюллера (с низким давлением).

Приводится краткое описание повторного определения электронного захвата β+-излучения в ветвях распада в Co$^{58}$. Этот метод включает абсолютный β+-счет и метод, основанный на совпадениях.
Algunas observaciones acerca de los progresos realizados en la calibración de núcleos que se desintegran por captura electrónica

El autor expone brevemente la medición de la actividad de núcleos de bajo número atómico que se desintegran por captura electrónica, empleando el método de coincidencias X-γ 4π y un contador proporcional 4π a presión (hasta 4 atmósferas). También describe un nuevo contador proporcional 4π de acero. Este aparato se ha proyectado para trabajar a presiones de hasta unas 40 atmósferas. Compara los resultados obtenidos con este contador y con el de menor presión en los núcleos de bajo número atómico.

El autor describe seguidamente la medición de la actividad de núcleos de número atómico más elevado. Se han hecho experimentos con el contador de alta presión para la determinación de rayos X y con un contador de centelleo 4π provisto de centelleador de NaI y de material plástico. Se ha procurado calibrar las muestras utilizando un contador de centelleo líquido. El autor describe la determinación de la razón numérica entre los rayos X de la capa K y los de la capa L y la medición de los rayos X-K acompañados por rayos X-L. En el caso de los núcleos de elevado número atómico que se desintegran por captura electrónica, la determinación exacta de estas cantidades permite medir su actividad por recuento de los rayos X-L con un contador 4π, método que no ofrece mayores dificultades. En el trabajo se describe también la medición de los electrones de Auger emitidos por los núcleos de elevado número atómico, empleando para ello un contador Geiger-Müller 4π clásico (de baja presión).

El autor expone brevemente una nueva determinación de la razón de bifurcación captura electrónica/β+ en el Co60. Este método exige el recuento absoluto de las partículas β+ y el empleo de técnicas de coincidencias.

(a) Low-Z Nuclides

Until quite recently, the standardization of orbital electron capture nuclides at AERE was performed in the manner described in a previous publication by the author (3).

Very briefly, the method for Cr51, Mn54 and Zn65 used the X-γ coincidence technique since a knowledge of counter efficiency and X-ray fluorescence yield is not required. The K X-rays were detected in a small 4π proportional counter filled with 90% argon — 10% methane to a pressure of approximately 1 atmosphere. K X-ray measurement were also made in the 4π counter at elevated pressures; up to 4 atmospheres total filling pressure. It was found that the logarithm of the X-ray count rate, plotted as a function of inverse pressure, lay very closely on a straight line. Extrapolation of this straight line to 1/p = 0 was taken as a measure of the total number of K X-rays emitted. When this figure was divided by the non-relativistic K-fluorescence yield, ωK calculated by BURHOP (1), a total disintegration rate was obtained which agreed to within 2% or so with the coincidence measurements. Since this agreement was reasonable, a similar technique was employed to standardize Fe55.

There is no strictly theoretical justification for projecting the straight portion of the pressure graph back to infinite pressure and the results of CAMPION (4) using a 4π counter operating at much higher pressures than ours suggested that we should investigate the error introduced in the extrapolation. In addition it had been suggested that the figure for ωK which we used was too low by several per cent.
Accordingly, a steel proportional counter was constructed. This consisted of two D-shaped cylinders cemented together with a source plate sliding...
between them. The anode wires were coaxial and the minimum path-length in the counter was 3 cm. This counter was placed in a steel pressure vessel capable of withstanding approximately 1000 psi. The E. H. T. leads, which also carry the output pulses, were fed into the pressure vessel through spark plugs. 90% argon — 10% methane mixture was used up to a pressure of 3 psi. above atmospheric pressure, thereafter, tank argon was admitted into the counter to raise the pressure further. The counter has worked satisfactorily up to pressures of 200 psi. with Fe\textsuperscript{55} at which pressure the operating voltage is about 4500 volts (figure 1).

Sources of Fe\textsuperscript{55} were counted in the small $4\pi$ counter (up to 44 psi. total pressure). The familiar straight line plot from the small $4\pi$ counter was obtained while a “pressure plateau” was observed with the steel counter. (It was noticed in this experiment, and in subsequent experiments where 100% X-ray efficiency was obtained that further increase in pressure resulted in a falling off in count rate. This applied to measurements with argon, krypton and xenon as the filling gas.) The extrapolated X-ray count from the small counter was approximately 12% higher than the corresponding count rate on the steel counter plateau and it appears almost certain that the agreement between X-ray coincidence counting and the extrapolated count method was fortuitous. Using the recent figure of 0.283 for the K-fluorescence yield of manganese \cite{5}, which figure agrees with the semiempirical figure from BURHOP \cite{1}, to correct the figure from the plateau, restores the total disintegration rate to its former figure. Similar investigations using Cr\textsuperscript{51}, Mn\textsuperscript{54} and Zn\textsuperscript{65} have not yet been completed. A pressure plateau obtained in the small counter using 90% Kr — 10% CH\textsubscript{4} gave a K X-ray emission rate which agreed with that from the steel counter (A — CH\textsubscript{4} filling) to within 0.5% (figure 2).
(b) Medium-Z Nuclides

The standardization of medium-Z nuclides which decay by 100% electron capture and which emit no γ-rays has also been considered. Such a nuclide is Cs\(^{131}\). This isotope emits K X-rays of approximately 30 keV and L X-rays of approximately 4 keV. In principle, it should be possible to determine the number of emitted L X-rays by 4π gas counting and to determine the total number of K X-rays emitted by 4π scintillation counting. Gas counting the K X-rays in the present equipment was not pursued since the efficiency was well below 100%.

A further approach in the case of Cs\(^{131}\) is to determine the number of K-L X-ray coincidences which arise when an L vacancy is left following a K process. If there are N disintegrations per unit time, of which a fraction \(a\) proceed by K capture and \((1-a)\) by L capture and if further a fraction \(p_{KL}\) of the K processes lead to an L vacancy, then we have the following relations.

\[
\text{Count rate in a K X-ray counter, } n_K = N \times \omega_K \, E_K
\]
\[
\text{Count rate in an L X-ray counter, } n_L = N \times (1-a) \times \omega_L \, E_L + N \times n_{KL} \times \omega_L \, E_L
\]

entre les rayons X K et X L émis, et comment il a mesuré le nombre de rayons

where the second term is the contribution to L X-rays following K capture.

The coincidence rate between two such counters, \(n_C\) may be expressed as:

\[
\text{The coincidence rate between two such counters, } n_C = N \times n_{KL} \times \omega_L \times E_L
\]

From these equations it is easy to show that:

\[
\frac{n_K \, n_L}{n_C} = N \left\{ d + \frac{(1-a)}{n_{KL}} \right\}
\]

Hence, knowledge of the K/L capture ratio and the \(K_\alpha/(K_\alpha + K_\beta)\) ratio enables a coincidence measurement to give the disintegration rate, N, without knowledge of the fluorescence yields.

A series of coincidence measurements were made between the two halves of the steel 4π counter. A Cs\(^{131}\) source was prepared on 260 \(\mu\)g/cm\(^2\) aluminium foil and was sandwiched between two aluminium foils of 1.37 mg/cm\(^2\) thickness. This extra foil reduces the number of K Auger electrons which would otherwise be counted. Single-channel, pulse-height analysers select K X-rays only from one half the counter and, with a narrow channel width, select a fraction of the L X-rays from the other half. A correction has to be made to the L X-ray count rate and to the coincidence count rate to subtract those counts due to K Auger electrons and other radiation occurring in the window. The magnitude of this correction is easily determined by scanning the single channel and coincidence spectrum beforehand when the L X-ray peak can clearly be seen superimposed on the general background of electron counts etc. (figure 3).

We currently use the figures of ROBINSON et al. \(^{(2)}\) for \(n_{KL}\) and \(P_L/P_K\), the ratio of probability of L capture to K capture.
In addition to these measurements, we have attempted to measure directly, the number of K X-rays and the number of L X-rays. The number of L X-rays was measured directly, in the presence of K radiation, in the steel 4\pi counter. The source was sandwiched between two layers of 520 \mu g/cm^2 aluminium. The calculated penetration through this aluminium is 64%. The number of L X-rays counted was determined from the difference in total counts with the discriminator bias set below and above the L peak, multiplied by the ratio of
the area of the L peak to the total area under the L peak (i.e. including the K contribution between the discriminator levels) (Figure 4). A further addition must be made for those L X-rays which are detected in coincidence with K radiation and hence appear with a pulse height different from the true L X-ray pulse-height.

The K X-rays were counted in a 4π NaI(Tl) scintillation counter arrangement. The crystals were 38 mm diameter and 3 mm thick and were covered with 6.8 mg/cm² aluminium. The source was sandwiched between aluminium foils such that the overall density of absorbing material between the source and crystal was 8.2 mg/cm². The calculated penetration of xenon K X-rays through such a sandwich is 95.4%. The absorption of the K X-rays in the crystal should be essentially 100%. The corrected count rate showed a plateau as a function of bias from which K X-ray emission rate was deduced (figure 5). The ratio of this K X-ray count rate to the total disintegration rate, the K-fluorescence yield, was 0.87. This result is in agreement with recently published figures for xenon of 0.85 (5) and 0.87 (2). Our estimate of the ratio of L X-ray intensity to K X-ray intensity was 0.073 which is in disagreement with the figure of 0.121 quoted by ROBINSON et al. (2).

The number of K Auger electrons emitted by a Cs¹³¹ source sandwiched between 260 μg/cm² aluminium was determined in a 4π Geiger counter filled with 10 cm argon-alcohol. A correction is applied for L X-ray efficiency but no correction has been applied for foil absorption of the K Auger electrons. This measurement, together with that of the K X-rays yields an ωₖ of 0.86, though this agreement may be fortuitous.

Unfortunately, measurements with a Cs¹³¹ source mounted between two halves of a plastic scintillator and measurements incorporating the Cs¹³¹ solution in a liquid scintillator have not given reproducible results up to the present time.

(c) Electron-capture/β⁺ Ratio in Co⁵⁸

In connection with the standardization of some Co⁵⁸ sources some time ago we measured the EC/β⁺ ratio for this nuclide. The method was similar to that published recently by HARMER et al. (7).
The decay scheme of Co\textsuperscript{58} is well known, and is shown in figure 6. The overall strength of a source was determined by the $4 \pi \beta$-$\gamma$ coincidence technique with the $\gamma$ counting channel set to admit the 0.81 MeV photo-peak.

![Figure 6](image)

All the positrons are accompanied by a 0.81 MeV $\gamma$-ray. The number of positrons was determined by counting the number of triple coincidences between the positrons and their two annihilation quanta. The Co\textsuperscript{58} source was sandwiched between two plastic scintillators which were thick enough to annihilate all the positrons. Outside this counting assembly were two NaI scintillation counters in coincidence. They could be set at 180° or 90° with respect to one another with the source as origin (figure 7).

![Figure 7](image)
If a fraction $\epsilon$ of the disintegrations proceed by positron emission, then the count rate in the positron detector is given by $n_\beta = N\epsilon E\beta$ where $E\beta$ is the overall efficiency for positron detection. The count rate in each of the matched $\gamma$ detectors is given by $n_\gamma = N\epsilon E_{511}\Omega + N\epsilon E_{810}\Omega$ where $E_{511}$, $E_{810}$ and $\Omega$ are the intrinsic efficiencies for 511 keV and 810 keV $\gamma$-rays and the solid angle subtended count rates at 180° and 90° is given by $n_{\gamma\gamma\text{diff}} = N\epsilon E_{511}.E_{511}\Omega$ and the difference between triple coincidences at 180° and 90° is given by $N\epsilon E_{511}.E_{511}\Omega$. We see immediately that the ratio $n_\gamma / n_\text{triple}$ is simply $N\epsilon$. The triple coincidence rates encountered in practice are rather low, approximately 50 per minute so that good stability of the apparatus is essential. The ratio of $N\epsilon/N$ obtained by the two methods was 14.6% which is in good agreement with other published values (6).

REFERENCES

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II. 3. 2. THE PROPORTIONAL COUNTER — SOME ASPECTS OF OPERATION

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Abstract

1. Work on a counter system designed for electron capture studies has shown that the diameter of the anode wire in a high-pressure proportional counter can have an important influence on the energy resolution obtained in measurements of low-energy radiations. In a counter working at a pressure of 12 atmospheres the width of the pulse distribution due to 13.5 keV K-capture radiations could be changed from 65% to 17% by changing the wire diameter from 0.0076 mm to 0.0025 mm. This phenomenon is interpreted as an effect of positive ion space charge in the avalanche. For a given gas gain and primary radiation the density of positive ions in the avalanche increases rapidly with pressure of the filling gas, and when the accompanying electric fields are large enough to affect the last stages of the multiplication process the energy resolution of the counter deteriorates. Decrease in diameter of the anode wire increases the externally applied field in the region where multiplication takes place, thus tending to counteract the effect of space charge. Use of 0.0025 mm anode wires has proved satisfactory in new measurements of L/K capture ratios made with a multiwire counter system working at pressures up to 12 atmospheres. The results obtained for L/K capture ratios of Ge$^{71}$ and Kr$^{79}$ are 0.115 and 0.112 respectively. The same counting system has also been used in a search for M capture in these two isotopes. The low energy of the M capture radiation, 180 eV in Ge$^{71}$ and 260 eV in Kr$^{79}$, and its low intensity relative to that of K capture, makes accurate measurement difficult; but peaks attributable to M capture were observed in pulse spectra of both sources.

2. Low-Background Counting—Recent observations made on a combination of proportional counter and scintillation counters (operating as shields) show that there are some real difficulties in devising a relatively simple system of this kind. The performance is inherently very satisfactory but practical matters such as long-term gas purity are difficult to overcome.

3. Studies of Electron Attachment — An approach to systematic observations in gases and mixtures of moment in proportional counting is described. It is hoped that such measurements as are effected with this arrangement will be valuable in enlarging the field of application of the counters.

Le compteur proportionnel — Ses caractéristiques de fonctionnement

1. Les travaux consacrés à la mise au point d'un dispositif de comptage destiné à l'étude de la capture électronique ont montré que le diamètre du fil constituant l'anode dans un compteur proportionnel à haute pression peut avoir une influence importante sur le pouvoir de résolution en énergie obtenu dans les mesures de rayonnements de faible énergie. Dans un compteur travaillant sous une pression de 12 atmosphères, l'amplitude de la répartition des impulsions dues aux rayonnements de capture K de 13,5 keV a pu être ramenée de 65% à 17%, en réduisant de 0,0076 mm à 0,0025 mm le diamètre du fil. Ce phénomène est interprété comme constituant un effet de la charge spatiale en ions positifs dans l'avalanche. Pour un gain et un rayonnement primaire...
donnés, la densité des ions positifs dans l’avalanche s’accroît rapidement avec la pression du gaz de remplissage, et lorsque les champs électriques d’accompagne-
ment sont assez grands pour affecter les derniers stades du processus de multi-
plication, le pouvoir de résolution en énergie du compteur diminue. Toute
diminution du diamètre de l’anode entraîne un accroissement du champ appliqué
extérieurement, dans la région où se produit la multiplication, ce qui tend à
contrecarrer l’effet de la charge spatiale. L’emploi de fils d’un diamètre de
0,0025 mm a donné des résultats satisfaisants lors de nouvelles mesures des
rapports de capture L/K effectuées au moyen d’un dispositif de comptage à fils
multiples, travaillant sous des pressions pouvant atteindre 12 atmosphères.
Les résultats obtenus pour les rapports de capture L/K du Ge$^{71}$ et du Kr$^{79}$ sont
de 0,115 et 0,112 respectivement. On a utilisé le même dispositif de comptage
pour étudier la capture M dans ces deux isotopes. La faible énergie du rayonne-
ment de capture M, soit 180 eV pour le Ge$^{71}$ et 260 eV pour le Kr$^{79}$, ainsi que
sa faible intensité comparée à celle de la capture K, rendent difficiles les mesures
précises; cependant, des pics imputables à la capture M ont été observés dans
les spectres d’impulsions des deux sources.

2. Comptage d’une activité de fond peu élevée — Des observations récentes
faites avec un dispositif comprenant un compteur proportionnel et des comp-
teurs à scintillateurs (ces derniers agissant comme écrans protecteurs) montrent
que la mise au point d’un dispositif relativement simple de ce genre présente de
sérieuses difficultés. Le fonctionnement est très satisfaisant en soi, mais certains
problèmes pratiques, tels que le maintien d’un gaz à l’état pur pendant une
longue période, sont difficiles à résoudre.

3. Etudes sur l’accumulation d’électrons — On décrit une méthode
d’observation systématique portant sur les gaz et mélanges importants pour le
comptage proportionnel. On espère que les mesures effectuées grâce au dispositif
correspondant s’avéreront utiles en permettant d’élargir le champ d’application
des compteurs.

Пропорциональный счетчик — некоторые аспекты работы

1. Работа над счетной системой, предназначенной для изучения захвата электронов, показала, что диаметр аноидного провода в пропорциональном
счетнике с высоким давлением может оказывать большое влияние на
разрешение по энергии, получаемое при измерениях радиации низкой
энергии. В счетнике, работающем при давлении в 12 атмосфер, ширина
распределения импульсов при радиации в 13,5 кэв K-захвата может
быть изменена от 65% до 17% посредством изменения диаметра провода от
0,0076 до 0,0025 миллиметра. Это явление объясняется как действие
положительного пространственного заряда иона в лавине. Для определен-
ного усиления газа и основной радиации плотность положительных ионов
в лавине быстро увеличивается вместе с давлением наполняющего газа, и
когда сопутствующие электрические поля становятся достаточно большими,
чтобы повлиять на последние стадии процесса умножения, разрешение по
энергии счетчика ухудшается. Уменьшение диаметра аноидного провода
увеличивает появляющееся извне поле в том месте, где происходит
умножение, и тем самым стремится противодействовать действию простран-
ственного заряда. Использование 0,0025-миллиметровых аноидных проводов
оказалось удовлетворительным при новых измерениях коэффициента
захвата L/K, проводимого с многопроводной счетной системой с давлением
до 12 атмосфер. Результаты, полученные для коэффициента захвата L/K с Ge$^{71}$ и Kr$^{79}$ равны 0,115 и 0,112 соответственно. Такие счетная система использовалась при исследовании M-захвата в этих двух изотопах. Низкая энергия радиации M-захвата, 180 эв в Ge$^{71}$, 260 эв в Kr$^{79}$ и ее низкая интенсивность по сравнению с интенсивностью K-захвата затрудняют точное измерение; но высшие точки благодаря M-захвату наблюдались в импульсных спектрах обоих источников.

II. Счетчик с низким фоном. — Последние наблюдения, проведенные на комбинации пропорционального и сцинтилляционного счетчиков (действующих в качестве экранов), показывают, что имеются некоторые реальные трудности в создании простой системы такого рода. Действие ее в принципе весьма удовлетворительно, но трудно разрешить такие практические вопросы, как долговременное сохранение чистоты газа.

III. Изучение электронного приспособления. — Описывается подход к проблеме систематических наблюдений момента в пропорциональном исчислении в газах и смесях. Надеются, что такие измерения, на которые оказывает воздействие это устройство, будут полезны для расширения пределов применения счетчиков.

El contador proporcional — Sus características de funcionamiento.

1. Los trabajos de perfeccionamiento de un dispositivo de recuento destinado al estudio de la captura electrónica demuestran que el diámetro del alambre anódico de un contador proporcional de elevada presión puede ejercer una influencia considerable sobre el poder de resolución energética obtenido en la medición de radiaciones de baja energía. En un contador que trabaje a la presión de 12 atmósferas, la amplitud de la distribución de los impulsos debidos a radiaciones de captura K de 13,5 keV, puede reducirse de 65 a 17% sustituyendo el alambre de 0,0076 mm de diámetro por otro de 0,0025 mm. Este fenómeno se interpreta como un efecto de la carga espacial de los iones positivos en la avalancha. Para una amplificación gaseosa y una radiación primaria determinadas, la densidad de los iones positivos en la avalancha aumenta rápidamente con la presión del gas contenido en el contador, y cuando los campos eléctricos concomitantes tienen suficiente intensidad para afectar las últimas fases del proceso de multiplicación, el poder de resolución del contador disminuye. La reducción del diámetro del alambre anódico trae aparejado un aumento del campo externo en la zona donde se verifica la multiplicación, tendiendo así a contrarrestar el efecto de la carga espacial, pero el empleo de alambres anódicos de 0,0025 mm ha dado resultados satisfactorios en nuevas mediciones de las razones de captura L/K efectuadas con un contador de hilos múltiples funcionando a presiones de hasta 12 atmósferas. Los resultados obtenidos para la razón de captura L/K del Ge$^{71}$ y del Kr$^{79}$ son 0,115 y 0,112 respectivamente. El mismo sistema de recuento se ha empleado para investigar una posible captura M en estos dos isótopos. La baja energía de la radiación emitida por captura M, a saber 180 eV en el Ge$^{71}$ y 260 eV en el Kr$^{79}$, así como su reducida intensidad en relación con la correspondiente a la captura K, dificultan una medición exacta; sin embargo, en los espectros de impulsos de ambas fuentes se han observado máximos que pueden atribuirse a la captura M.

2. Recuento de bajas actividades de fondo. — Observaciones recientes efectuadas con un dispositivo formado por un contador proporcional y conta-
dores de centelleo (que funcionan como pantalla protectora) revelan que la construcción de un dispositivo relativamente sencillo de este tipo ofrece ciertas dificultades. El funcionamiento es de por sí muy satisfactorio, pero algunos problemas prácticos, tales como el mantenimiento del gas al estado puro durante un tiempo largo, son difíciles de resolver.

3. Estudios sobre acumulación de electrones. — El autor describe un procedimiento de observación sistemática de las propiedades de los gases y mezclas utilizadas en el recuento proporcional. Cabe esperar que las mediciones efectuadas con este dispositivo permitan ampliar el campo de aplicación de los contadores.

The proportional counter is undoubtedly a most powerful tool in the study of nuclear radiations. Its applicability over a wide range of modern physics has grown rapidly ever since its introduction in modern form for the energy determination of lightly-ionising particles in 1948 (1), (2). In the following pages it is hoped to present the main features of some recent studies which extend its usefulness in several directions.

I. Use at High Pressures

The counter has been shown to possess on the whole rather satisfactory energy resolution for ionising particles whose tracks are confined to the active volume of the counter. Some study of the factors determining the resolution has been made. Thus approximately it is true to say that the peak width varies with the value of \sqrt{n} where n is the number of ion-pairs formed along the initial track. Likewise the multiplying collisions vary statistically in number and again in proportion to \sqrt{n}. These components of the peak width were shown experimentally, in normal conditions of operation, to be about equal and the result could be expressed as \( n \pm \sqrt{\frac{2}{3} n + \frac{2}{3} n} \). However as n increased it was found that the energy resolution did not improve relatively as much as was to be expected on this view. The work at higher energies, higher n values, involves the use of higher pressures and careful studies did not readily reveal any answer to this problem.

Recently Drever and Dougan at Glasgow, in working with radioactive gases and vapours, have made measurements on a most interesting phenomenon which may have bearing on the above. The research involved the measurement of K, L and M radiations emitted mainly in capture processes and the special counter assembly was in fact designed to achieve close to 100% absorption of the X-radiation. The important features of the technique have been described (3), (4). Essentially a considerably modified form of a novel low-background counter (4), (5) was devised. Here a wire-bounded counter lies within a shielding cage of wire-bounded counters. No materials save the wires separate any one counter from its neighbours. This arrangement is such that radioactive gaseous constituents of the filling gas or vapour may be accurately measured both in intensity and energy and only relatively small corrections need to be applied. For example, K radiation arising in the innermost axial counter may escape but provided that it is detected in the surrounding shielding assembly, the correct
intensity is nevertheless measured by the central counter. This is true whatever fraction is stopped in the axial one since the fraction escaping is counter-balanced by the fraction of the counting rate due to radiations having their origin in the shielding counters but which are actually absorbed in the axial one.

There is of course an end correction but with long counters operated at high pressures the approximation to 100% detection in the shield can be made rather close. This result depends on the use of satisfactory amplifying and anti-coincidence circuits with the system. The ring counter surrounding the main proportional counter is connected in anti-coincidence with it. If for example the source emits K and L X-rays and the bias on the two counter systems is set suitably, measurements of L capture can be made free from effects due to escape of K X-rays emitted by the gas source. In other words, if L X-radiation detected in the inner counter is true L-capture radiation no K X-rays are observed in coincidence in the ring, while if the L X-radiation is merely subsequent to K capture the K capture quanta are observed in the ring and used to cancel out automatically the L radiation arising incidentally. Hence true values of the intensity of L capture can be deduced from the spectra observed on the kicksorter. As we have said this work depends on the successful use of high gas pressures and as the radiations are usually of very low energy little success was achieved at first. Operating mainly at 6
and 12 atmospheres of argon (with 15 cm of methane) Drever and Dougan found that the counters had entirely inadequate resolution when the wire diameter was the rather usual 4 mil or so in diameter. An example of this is shown in figure 1 a for a 3 mil wire and a pressure of 12 atmospheres of argon with 15 cm of methane. On the other hand, with reduced wire diameter and otherwise identical conditions, resolution was greatly improved as shown in figure 1 b. Examples of the spectra taken with a little krypton (Kr\textsuperscript{79}) in

![Figure 1](image)

Figure 1

L and M peaks for Kr\textsuperscript{79} and Ge\textsuperscript{71}.

Whereas the measurements are entirely impossible at a wire diameter of 4 mil the resolution is reasonably satisfactory with wire of diameter 1 mil at pressures of 12 atmospheres or even more. It should be noted that in the observing of such low-energy radiation many extremely large pulses are present and the amplifier must be adequately paralysed for times up to about 5 msec. The peak of M radiation of Ge\textsuperscript{71} is the lowest energy yet observed in nuclear spectroscopy and it is found to be displaced to lower energies than might be anticipated. This is probably associated with the nature of the mechanism of ionisation in gases at these reduced energies. The rising count at still lower energies is spurious and dependent on the paralysis circuit.

The work described here has led directly to experimental determinations of L/K and M/L ratios of rather good accuracy and the agreement with recent theoretical studies is closer than before. There seems to be a definite systematic difference for the L/K ratio of about 10% between the two. Further information on this problem can be found elsewhere and is not relevant to the main discussion here. However, we may note in passing that the L/K capture rations measured were for K\textsuperscript{79} 0.108 ± 0.006 and for Ge\textsuperscript{71} 0.115 ± 0.006, reasonably near to the theoretical values of 0.102 and 0.106. The previous

![Figure 2](image)

Figure 2

L and M peaks for Kr\textsuperscript{79} and Ge\textsuperscript{71}.
The experimental value for Kr\textsuperscript{79} was 0.206. Regarding the M peaks shown in the figure it is of note that the expected energies are only 180 eV and 260 eV while the expected intensities are about 2\% of the K-capture intensities. Nevertheless, the counter shows the peaks in measurable form even though there appears to be an energy displacement. A similar displacement has been observed again by Drever and Dougan for A\textsuperscript{37}. The expected M/L ratios of 0.15 and 0.16 for Ge\textsuperscript{71} and Kr\textsuperscript{79} respectively, are agreeable with the results within the rather approximate accuracy of the actual measurements.

We should now like to say something about the nature of the effect of wire diameter at higher pressures. The effect is of considerable practical importance and it is desirable that it should be understood. The first suggestion that arose was that it depended on the space charge at the wire. For a given gas gain and primary radiation the density of the positive ions in the avalanche must increase with the pressure of the gas. When the accompanying space charge fields become large enough to affect the multiplication process the energy resolution of the counter deteriorates. At reduced wire diameter we have higher field and field gradient in the very important region where multiplication takes place, that is in the immediate vicinity of the wire. Changes in the field strength produced by space charge might be therefore less serious the greater the field gradient. If the last stages of multiplication are reduced by the positive ion space charge we would expect gas gain to have a large influence on the resolution. With the gas gain and amplifier gain varied by a factor of 10 or more and the overall gain maintained constant no change in the results for thin or thick wires were observed. A great deal of fine work on the effects of scratches and other irregularities has been done by D. West and he pointed out to Drever that such irregularities might be the cause of field disturbances similar to those expected from space charge; assuming the irregularities are of the same relative magnitude. Very recent work of Drever has gone some way to confirm this view. A small localized source of monoenergetic X-rays was moved over the inside of the counter while it operated. Ge\textsuperscript{71} was used in solid form so that the X-rays, largely of energy 9.2 keV, had an absorption half-thickness on only 3 mm in the argon-methane mixture at the pressure of 12 atmospheres. With a 3 mil wire the mean pulse-height and the resolving power depended strongly on the position of the source. Both movements parallel to the axis and around the circumference caused variations in pulse height of up to 50\%. Good resolution was obtained with the source in certain selected positions. This work tends to indicate that the main field disturbances causing poor resolution when gas sources are involved are probably due to irregularities in the anode wires rather than to space charge effects.

It seems reasonable to deduce from this current work, and particularly from the measurements made with the source moving round the circumference, that with the thicker wires the avalanche is well localized and confined to an angle of less than about 45\% at the wire surface. The expectation is that with 1 mil wire the avalanche spreads around a greater fraction of the circumference. Such spreading of the avalanche together with the greater field gradients may tend to reduce considerably the effects of irregularities in the case of the very thin wire. This conclusion is not yet based on experiment and for the present all that can be asserted is that it is better in practice to use a thin wire rather than a thick one at high pressures. It remains to be
seen if this is still at all true for uniform accurately circular wires free of irregularities. The most recent observations indicate that other wire materials, notably platinum, are to be preferred to tungsten, giving better resolution at the same wire diameter.

II. Electron Attachment

Counter Measurements

The resolving power as a function of wire diameter at elevated pressures has been discussed. The attachment of electrons to neutral atoms and molecules can have serious effects and there is relatively little available data peculiarly applicable to proportional counters. It was in fact first suspected in the work described above that attachment was involved to some extent but subsidiary experiments with simple counters of different diameters eliminated this possible cause. Moreover calcium and hot copper purification did not produce noticeable effects. It is true to say however that it is at high pressure that we expect the presence of planned or accidental gaseous additives to the standard mixtures to be most serious. For such reasons Dr. F. Morgan's group at AWRE have started work in this field and some systematic examination of the role of electron attachment on the behaviour of proportional counters has been initiated by Dr. H. W. Wilson. The programme falls into two main parts:

a) the measurement of plateau gas gain and resolution as functions of the pressure of the additive and of the main gas,
b) the study of attachment in linear electric fields so that attachment in terms of \( E/P \) can be measured.

The electrons in the track of an ionising particle move towards the wire at first in relatively low fields. If some of the electrons are attached to form negative ions these drift much more slowly towards the wire and there is no contribution from these to the pulse amplitude. Thus the pulse amplitude for a given counter is usually reduced when attachment is present. However, the resolving power appears to deteriorate much more rapidly than one might expect from the reduction in \( n \), here regarded as the effective number of electrons which reach the multiplying region round the wires as can be seen by examining some of the curves obtained. This observation refers to the case of rather broadbeam irradiation of the counter with primary tracks distributed through a rather large volume of gas. The question arises as to whether the important period of occurrence of attachment occurs during the time of drift of electrons towards the wire. We are of course neglecting the problems presented by later arrival of negative ions. Experiments performed with a directed beam of electrons, at angles between zero and 90° to the wire, should make our conclusions more precise in this matter and such a study is under way. The time of arrival of electrons in the track at the multiplying region round the wire will then be variable and more precise observations will be possible.

The practical problems presented by electron attachment are serious. Many gases, e.g. Ni(CO)₄ have to be used in gaseous form to obtain the optimum results yet attachment is extremely marked and not as yet explicable on theoretical grounds. Again ‘poisoning’ occurs as gases or vapours are released in small amounts from the walls. There are anomalies here as for example with argon + methane (90/10 by pressure) satisfactory operation may
be maintained for several days while with argon + methane (30/70 by pressure) the performance can be hopelessly poor in less than one day.

In measurements of class a) above a proportional tube of 40 cm length and 10 cm diameter with a wire of 2 mil diameter was employed. With a filling of argon + 10% methane the resolution of the 9.2 keV peak of Ge$^{71}$ was 14% and the plateau length was 300 volts. A window of 5 mg/cm$^2$ thickness was used as the X-ray port. As examples of the results we may refer to the work on oxygen, hydrogen and carbon tetrachloride.

![Figure 3a](image)

The form of plateau curves for argon + methane at various partial pressures of O$_2$.

![Figure 3b](image)

The resolution as a function of added O$_2$. 

361
Oxygen. In figures 3 a and 3 b we show the effect on the plateau (and pulse amplitude incidentally) of adding increasing amounts of oxygen to the standard mixture and the deterioration of the resolution of the counter for the 9.2 keV radiation from Ge\textsuperscript{71} with increasing oxygen content. It is impossible to estimate the width of the peak at half-height when the partial pressure of \(O_2\) rises to about 2 mm of Hg. The spectrum shows a broad continuum of pulses of amplitude less than the value of 9.2 keV at the peak.

Hydrogen. In figures 4 a and 4 b we show similar curves to those of figures 3 a and 3 b. With increasing hydrogen content the plateau moves to higher voltages and the length gradually reduces. The resolution deteriorates at the same time but even at a partial pressure of 15 cm Hg the 9.2 keV peak of Ge\textsuperscript{71} is still well resolved. The resolving power as a function of pressure is not yet sufficiently well determined.

![Figure 4 a](image)
Plateau curves for added H\(_2\) in 76 cm of A + CH\(_4\).

Carbon Tetrachloride. This gas showed very marked effects. No proper counting could be done at a partial pressure of 1 mm of Hg and even at 0.5 mm of Hg the counting rate was reduced drastically and the peak at 9.2 keV was not at all resolved. The nature of the effect on the counting rate of the addition of 0.5 mm Hg of CCl\(_4\) is shown in the following figures:

362
Direct Measurement

There are two main types of electron attachment experiment: firstly those in which the negative ions are formed by interaction of electrons and gas molecules in one chamber and then transferred into a high-vacuum magnetic or electric analyser and, secondly, those in which all operations occur in one chamber. The apparatus being built is intended for the second type.

A parallel plate condenser is contained in a glass tube which may be pumped out and filled with the gas to be tested at any pressure up to atmospheric. One of the plates is a quartz disc with a thin coating of silver from which photo-electrons are liberated by short pulses of ultra-violet radiation from behind. This plate is connected to a variable high-voltage supply of negative polarity. The other plate which is moveable to alter the condenser thickness is surrounded by a guard ring at the same potential and is earthed through a resistance. The potential difference across the resistance is fed into an amplifier and cathode-ray oscilloscope.

When electrons are ejected from the photo-cathode they accelerate and reach a terminal "drift-velocity" due to collisions with gas molecules. The current pulse in the external circuit as shown by the oscilloscope will then have a rise, depending on the production rate and acceleration of the electrons,
followed by a steady portion during the constant drift and a final fall depending again on the production of the electrons but also on their scatter by collision in transit.

However, if the gas molecules tend to take up electrons to form negative ions, the steady portion of the measured current-pulse will in fact be subject to an exponential decay caused by the loss of electrons plus a smaller exponential increase due to detachment of ions already formed plus a contribution due to the negative ions drifting much more slowly because of their large mass.

In this apparatus the condenser plate diameter (excluding guard ring) is 7.6 cm and the condenser thickness is variable from zero to about 15 cm. In order to check uniformity of field provision is made for ejecting only central or only peripheral electrons. The transit times of both should be equal.

The ultra-violet flash tube is hydrogen filled to a pressure of 100 mm Hg and its design is based on that of MALMBERG for a millimicrosecond light source. Its envelope is of quartz.

The flash tube is situated in an evacuated chamber. This reduces absorption of the ultra-violet and permits the tube to be brought close to the photocathode without high-tension breakdown.

It should be mentioned that an apparatus similar in principle but employing a pulsed flat beam of X-rays for electron production by ionisation of the gas has been used.

Other workers have used photo-electric sources but in rather different ways.

In figure 5 is a schematic diagram of the apparatus.

![Figure 5](image)

**Arrangement for attachment measurement in uniform field.**

**Measurements and Discussion**

It can be shown that if some electrons become attached to gas molecules during transit of the chamber the current falls off exponentially and is given by

\[ I = I_0 e^{-\alpha w_e t} \]

where \( \alpha \) is the electron attachment probability per unit drift distance in the field, \( w_e \) is the electron drift-velocity and \( t \) is the time of transit.

364
The current wave form is displayed on an oscilloscope upon which visual measurement of I, I0, and t can be made. Knowing t and the distance between the electrodes gives a measure of the electron drift velocity in the gas under investigation. Then α can be deduced from equation (1).

Errors in the measurement of 't' may be introduced by i) diffusion effects of the initial electron cloud, ii) the finite time taken to release the electron cloud from the photo-cathode, iii) the time constant of the electronic circuitry, and iv) the time taken for the photo-electrons to reach their terminal drift-velocity. To make these effects negligible compared with the transit time of the electrons (~ 1 μsec) a milli μsec ultra-violet light source is used and the RC constant for the chamber is made ~ 10⁻⁸ sec. Diffusion effects are removed by interpreting the time interval as the time between the mid-points of the leading and falling edges of the current-pulse waveform.

The distance travelled from the cathode before the photo-electrons attain their terminal drift-velocity characteristic of the applied field will be dependent on the gas pressure. Thus although this distance is expected to be only a fraction of a millimeter, a check can be made on the magnitude of the effect by varying the distance between the electrodes and keeping E/p constant. If the time taken for the electrons to reach a limiting velocity is negligible the calculated electron drift-velocity will be independent of the plate separation.

CHANIN et al. (10) have shown that for oxygen at low electron energies < 1 eV the three-body electron attachment process is predominant since a marked pressure effect occurs. Above 2 eV, the two-body process is prevalent.

CRAGGS et al. (11) have verified that this is the dissociation attachment process.

The probability of a two-body collision occurring between an electron and a neutral molecule (e.g. O₂ + e → 0 + O⁻) is proportional to the gas density, φ, whereas the probability of a three-body process involving the electron, the molecule and another molecule which stabilizes the reaction is dependent on φ². Hence α can be expressed in the form α = βφ + K²φ where β is the two-body attachment coefficient, and K is the three-body attachment coefficient.

It is planned to obtain measurements of α/p for different values of E/p (volts/cm/mm Hg) at different pressures for a wide range of pure gases, common vapours and mixtures. E/p values ≤ 20 at pressures from 0—760 mm Hg are possible with the apparatus described.

From the curves of α/p and E/p the values for β and K as functions of the average electron energy will be deduced.

The above work is being prosecuted as part of a programme of Bannister, Dunmur, Watt and Wilson at AWRE.

III. Low-Background Counting

A great deal of study of the problem of counting extremely weak sources has been prosecuted in recent years. We shall attempt in the following to outline some of the work at AWRE.

A new low-level laboratory has been constructed after considering various alternative approaches. The unit consists of a double-walled tank containing 6 ft of water all round and with a depth of 7 ft of water on top, with a floor of 2 ft of concrete and 3 inch of steel. The room within is circular in form.
with a diameter of 21 ft. The entrance is by a steel door of 1 ft thickness. Electrical leads are taken through curved channels to maintain shielding in all directions at 1 ft of steel or 6 ft of water.

Two forms of counter have received most attention. The first of these is the multi-wire type (4) (5) discussed above in connection with wire-diameter effects. This has proved remarkably easy to keep in good operation even with 1 mil wires and the performance can be excellent, in the region of 0.6 cpm per litre of active counting volume. Its rather low volume efficiency makes it less suitable for studies of certain sources such as C\textsuperscript{14} where important samples of interesting age may be involved and only a limited amount of material may consequently be available. Here the fact that much of the gas space is outwith the central counter is a drawback. Moreover in work at relatively modest pressure and moderate applied magnetic field there is a loss of counts due to coincidences between the inner and ring counters. Work on a small multi-wire counter of total volume 1.90 l, inner counter volume 0.725 l (geometric efficiency = 38.2%) showed that the second loss was very serious and an experiment was done with a thin internal wall between the ring and central counter. This consisted of 0.005 inch of cellulose coated with aluminium. The measured efficiency was 37.4 ± 0.5%. The background, with very little shielding, was 2.2 cpm without the wall and 4.1 cpm with it.

The most recent form of this counter has a stainless steel case of volume 24.6 l, giving an inner counter volume of 12.3 l and a geometrical efficiency of 50%. At only 1 atmosphere the counting loss is 5% when placed in a solenoid producing a field of 1750 gauss.

The second form of counter receiving attention at AWRE and at Glasgow is the proportional with walls of scintillating plastic. The dead volume problem discussed above is thus virtually eliminated and the escape problem at lower pressures can be solved by the choice of the electrical parameters of the anti-coincidence circuit. With C\textsuperscript{14} the pulses from the scintillation counter are

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Figure 6 a
Diagram of plastic phosphor/proportional counter assembly.
set in anti-coincidence only if they exceed the limiting energy of the C$^{14}$ β-rays, that is about 150 to 160 keV. The system has previously been described in principle by MOLJK et al. (12). A schematic drawing and photograph are shown in figure 6. The light from the plastic phosphor shield is collected by two 5 inch EMI photomultipliers. The wall and end thicknesses are $\frac{3}{8}$" and $\frac{1}{2}$" respectively. A cathode of nickel is vacuum deposited on the plastic. A one mil tungsten wire, 11 inches long is used and the proportional counter has a volume of 1.8 l. A purification system for the CO$_2$ gas reduces the volume efficiency to about 90%. With an argon-methane filling at 1 atmosphere a counting rate of 2.1 per minute over the C$^{14}$ spectrum has been measured, corresponding to about 1.2 cpm per litre of active volume. This was observed in a castle of 10 cm of lead and 10 cm of iron. The use of a purifying system has proved essential for the successful steady operation over periods of some days. This refers particularly to the measurement of the activity of CO$_2$ or CH$_4$ gas but even with these a simple hot copper wire purifier has been found satisfactory.

Electrical breakdown caused by tracking over the uncoated phosphor surface was eliminated by the use of thin PTFE discs. The phosphor is liable to craze and even to crack under pressure and optical coupling may be affected by a cloudiness developing at the surface. This can of course be removed by machining off the outermost layer.

Some Applications — decay of Rb$^{87}$ und Lu$^{176}$. It is of interest to remark here on one of the very important fields of low-background work — the measurement of the half-lives of important long-lived natural radioelements. Their role in geophysical dating studies is well appreciated. At AWRE McNair is involved in redetermining the half-lives of Br$^{87}$, Lu$^{176}$ and a few
other elements. For the former he has returned to the use of the 4 π thin source on thin film method (13) (14) introducing further refinements:

1. The thickness of the VYNS film and metal coating was reduced to about 10 μg/cm².
2. The counter was in an anti-coincidence Geiger ring and 8 in steel castle to reduce background and permit the use of thinner sources of lower activity.
3. Rb enriched in mass 87 (99.27%) was used to increase the specific activity.
4. The source was vacuum evaporated to give uniformity of thickness and its mass was determined by mass spectrometric isotope-dilution analysis using natural Rb as a spike.
5. The source area was increased to nearly 100 cm².

Features 1 and 5 introduced unexpected difficulties, for example the conductivity of the metal film was a very real problem.

The unusual combination of techniques in this experiment is of considerable interest. The half-life appears to be close to \(4.90 \times 10^{10}\) yr.

In respect of Lu\(^{176}\) which decays to Hf\(^{176}\) and possibly to Yb\(^{176}\) rather direct methods have been followed and these are designed to bypass much of the unknown in the nature of the decay scheme. The first method consisted of a \(\gamma\)-\(\gamma\) integration in a single crystal. The photo-fractions for two of the \(\gamma\)-rays in the cascade (\(\gamma_1\) and \(\gamma_2\)), or the fractions of all the \(\gamma\)-rays detected in the crystal which contribute to the photo-peaks, are required and they are in fact readily measurable. Several values obtained in this way ranged from \(3.58 \times 10^{10}\) to \(3.66 \times 10^{10}\) years with an average of \(3.61 \pm 0.05 \times 10^{10}\) years.

The second method, 4 π β counting with a thin source (200 μg/cm² on an aluminium backing of the same superficial mass) showed negligible loss by absorption and gave values of \((3.23 \pm 0.10) \times 10^{10}\) years and \((3.20 \pm 0.05) \times 10^{10}\) years. We believe that the true half-life lies within the bracket defined by the experiments but the first \(3.61 \times 10^{10}\) year is probably the more correct of the two since contamination by other radionuclides could affect the second. The value of \((3.6 \pm 0.1) \times 10^{10}\) year for Lu\(^{176}\) is at variance with the results of other observers (15) (16) (17) but not very different from the \(4.1 \times 10^{10}\) year given by McNAIR (18) on the basis of earlier work of DIXON et al. (19).

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Dr. Putman (United Kingdom):

I would like to put a question to Dr. Pate and Dr. Campion. We have found, in a series of comparisons of $4\pi$ Geiger-Müller counter measurements with $4\pi$ proportional-counter measurements, that the proportional counter measures consistently from 0.8—1% lower than the Geiger-Müller counter, or, looking at it another way, that the Geiger-Müller counter measures about 1% high. This effect appears to be almost independent of the maximum energy of the $\beta$ radiation measured and seems to be a function of the counter. It is not, we believe, an effect of spurious counts, since the Geiger-Müller counter plateaux are extrapolated to the threshold potential.

Dr. Pate (United States of America):

These data are clearly most important and it would be very helpful to have them published. It strikes me that, if they are correct, then they have only the two possible interpretations that Mr. Putman has mentioned. Whichever interpretation proves to be correct, there will be a group of workers who will be very disturbed indeed.

Dr. Grinberg (France) (Translated from French):

I just want to say that our results fully confirm Mr. Putman's. All our intercomparisons with the United Kingdom National Physical Laboratory have been based on our using a $4\pi$ Geiger-Müller counter and the NPL using a proportional counter. Results have been in systematic agreement within the limit of the accuracies claimed by the two laboratories — i.e., 2 or 3% depending on the radioisotope — but our results have consistently been slightly the higher of the two and fall exactly within the limits described by Mr. Putman in respect of measurements with the proportional counter.

Dr. Pate:

Since this conference is graced by the presence of an expert on proportional counting — I refer to Dr. Curran — would it be possible to ask him for his comments?

Dr. Curran (United Kingdom):

I am not really very surprised at this systematic difference between the Geiger-Müller absolute rate and the proportional absolute rate on the same source, because I am never very sure when a Geiger-Müller counter is counting an absolute rate, although I am sure when a proportional counter is counting an absolute rate because you can always check by an experiment with single electrons. Each single electron produced within the proportional counter is being observed. I know of no similar check with a Geiger-Müller counter and I would suspect the latter as being wrong when I got a difference.

Dr. Baptista (Portugal):

Has Dr. Putman tried to work with the same counter — i.e., with the same electrical arrangement — and source first in the proportional region, and then in the Geiger-Müller region to see if there is any difference? You obtained these data using two different counters, one in the Geiger-Müller region and the other one in the proportional region. Can you not do an experiment — I do not know if it could actually be performed — with the same counter and same source operating in the two regions consecutively?
Dr. Putman:

I think this could be done, but it would probably mean changing the centre wire of the counter — i.e., the anode wire — because it would not be working under optimum conditions in both cases.

Dr. Mann (United States of America):

We have actually carried out experiments with our internal gas counters (it is not the same system but it may be interesting to note) both in the Geiger region and the proportional region with the same original sample of C¹⁴, and although it is within the statistics of measurement and is small when you discriminate to zero discrimination in the proportional region, the Geiger-Müller results are systematically higher than those we obtained by proportional counting using the same counting system.

Dr. Campion (Canada):

I have a comment to make at this point. It is assumed, and I think fairly well established, that a Geiger-Müller counter will count a single electron. Dr. Curran was quite right when he said a proportional counter could be made to count a single electron, but in the usual counting systems which are set up in standardization laboratories this may or may not be so. In our case, one has to consider the counter and amplifier as a complete system and it is probably detecting just ten ion pairs. This will obviously make a difference in the counting rate between the Geiger counter and the proportional counter.

Dr. Curran:

I should like to come back a little to this question, because it has always baffled me of how you can be sure of where to set a Geiger-Müller counter on its plateau for 100% detection of particles. The proportional counter, on the other hand, with a suitable little light source, can always be shown to be counting from single electrons upwards when it is operating. It is much less likely to produce spurious pulses than the Geiger-Müller counter, and we know that in spite of refined quenching techniques and the like there is some finite possibility of spurious pulses affecting the Geiger-Müller return. So, for a really satisfactory operation in these conditions, one ought to have some sort of test procedure for the Geiger-Müller to know where you sit on the plateau for correct determination of the counting rate. You can get satisfactory determination of that position with the proportional counter, without actually looking at single electrons, just by passing some homogeneous radiation into the proportional counter, finding the pulse amplitude produced by that homogeneous radiation, and showing that a fraction of it — i.e., a fraction corresponding to about 1 electron — will probably be counted nearly 100% efficiently in the proportional case. Now you have at least two ways of telling with the proportional counter where you should set it to get 100%, and only 100%, detection. I do not think there is a comparable way with the Geiger.

Dr. Putman:

Could one combine the remarks of Dr. Curran with those of Dr. Campion and suggest that it may be that the proportional counter is to be preferred in absolute determinations but only if set up in the way suggested by Dr. Curran.

Dr. Curran:

Speaking for myself personally, I always set it up in the way I have described. I have tried a counter with either A³⁷ or Ge²¹, found the pulse size and reckoned that one electron could be detected. It is a very fast and easy way
of knowing where you should sit on your so-called plateau. I think you can get very close to correct absolute determinations, even when the plateau, for the many reasons that can affect plateaux, is not entirely satisfactory.

Dr. Campion:

This is very nice if you are dealing with monoenergetic radiation, but in dealing with β-emitting sources one has a range of pulse heights of at least 1 to 1000 and probably more; this is a very severe test for one's electronics to handle this, if one is counting one ion pair, and the possibility of double counting cannot be ruled out under these conditions. I think perhaps I could go into further discussion on this with Dr. Putman and Dr. Curran later.

Mr. Gasstrom (IAEA):

Dr. Mann, why do you introduce methane into your C\(^{14}\) counter as well as carbon dioxide? Would not carbon dioxide alone be satisfactory, as used by Prof. de Vries in his proportional counters at Groningen?

Dr. Mann:

It is just a question of purity. It is easier to do proportional counting with ordinary commercial methane, and so we prepare our active sample in the form of CO\(_2\) and then mix a little of this with a considerable quantity of methane for counting. If you wish to count with carbon dioxide, you have to purify it rather stringently.

Dr. Morgan (United Kingdom):

I should like to make an observation on a general point of technique which of itself might be only of minor importance. However, it has run through the discussion of the past few days. Several speakers have mentioned the use of micro-pipettes for the measurement of small volumes. These in our experience are not terribly accurate. They are fragile and expensive. Dr. Pate mentioned that carrier-free solutions deteriorate on standing in ampoules. Other speakers have mentioned the thickness of sources in 4\(\pi\) counting, which is much greater than would apparently be warranted by a knowledge of the chemical composition of the solution. We have in my laboratory avoided all these difficulties in a very simple manner. We have used small polythene ampoules for the dispensing by weight and the storage and transit of radioactive solutions. As regards the stability of these solutions, I would just like to quote two numerical examples. First, we have kept a solution with a cobalt concentration of approximately \(10^{-8}\) g per g of solution for a period of several months. The concentration of Co\(^{60}\) in this was determined in an interlaboratory comparison over this period, between ourselves and laboratories such as the United States National Bureau of Standards and Chalk River, and practically identical results have been obtained. This shows the solution is stable. Similarly, we have kept carrier solutions of U\(^{233}\) and U\(^{233}\) for periods of well over a year without any apparent change in their concentration. Finally, we have found that chemical operations in glass ampoules introduce impurities which can be easily observed and which are certainly not picked up from these small ampoules. They are hexagonal in cross-section and have a capacity of approximately 6 ml. If you can imagine a doll's feeding bottle made of polythene, that is precisely what they are.

Dr. Mann:

I would agree with Dr. Morgan, although I was one of the culprits for showing micro-pipettes on one of my slides, that we would never dream of
using them on routine work at the National Bureau of Standards. We use the pyknometer or the ultra-micro-burette, either of which is much more accurate. The pipettes are simply calibrated at delivery and you cannot deliver very precisely.

Secondly, I would like to ask Dr. Morgan whether there are any cases of liquid, I am not sure by which process, diffusing through the polythene bottles. We have had some such trouble with plastic bottles. We stored Sr\textsuperscript{90} once in a plastic bottle and there was about half the volume left when we came back. Fortunately all the Sr\textsuperscript{90} was there, but not all of the aqueous solution, I think it was.

**Dr. Morgan:**

If I understood Dr. Mann's comment correctly, he lost the liquid but not the Sr\textsuperscript{90}. This is a rather inexplicable phenomenon unless he had a pinhole in his ampoule and unless his strontium was in the form of a precipitate already. I can only repeat that in our experience the use of slightly acidified solutions of most common cationic materials has been that they are almost indefinitely stable.

**Dr. Mann:**

I am sorry, I am not talking about stability, actually I am talking about diffusion of liquids. I am not quite sure at the moment, but it is impossible to contain certain liquids in plastic containers. It may be I am thinking about alcohol or water, I am not quite sure at the moment. I was wondering if there was any difficulty about diffusion through the plastic container.

**Dr. Yaffe (Canada):**

Did Dr. Morgan try the same Co\textsuperscript{60} solution with the glass ampoule to check its stability? Co\textsuperscript{60} is one of the nuclides with which one rarely runs into difficulties. Has he done this with some of the other nuclides that are more difficult to keep?

**Dr. Morgan:**

It is obviously impossible within a reasonable time to study the behaviour of every chemical element in the many chemical forms which are possible. One would obviously not attempt, for example, to keep iodine, even as iodide, in such a solution. What we have done is to study the deterioration of carrier-free fission products, which cover a pretty wide region of the Periodic System, and to show that, over times of the order of days and weeks, we can get reproducible answers. If Dr. Yaffe wants to pursue this one further, perhaps we could discuss it individually outside.
II. 4. GAMMA COUNTING
(SESSION 5, 16 OCTOBER 1959)

II. 4. 1. THE PROOF SENSITIVITY OF WELL-TYPE CRYSTALS OF DIFFERENT SIZES WITH REGARD TO QUANTUM RADIATION FROM COMMON RADIONUCLIDES

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Abstract

Published works contain theoretical details of proof sensitivity of NaI crystals and some experimental data on proof sensitivity of well-type crystals. In order to assess the usefulness of a well-type crystal for a specific measuring problem, however, it is not sufficient to state the sensitivity of the detector. Only consideration of further factors, such as the nil effect of the arrangement, the time required for measuring and the degree of accuracy demanded can help to decide whether the measuring problem in question is capable of solution. A method for dealing with this inter-relationship is suggested.

While proof sensitivity is the main determining factor in deciding how far a problem can be solved, in calibrating radioactive sources more significance is attached to the actual number of impulses per unit of activity observed by the equipment available. Fundamentally, this factor depends on the particular nature of the electronic series connexion in the well-type crystal. For various combinations of equipment the sensitivities are given as determined experimentally for some commonly-used isotopes. The detailed discussion of errors includes the investigation of count losses in well-type crystal measurements. In addition, the dispersal width of the defined sensitivity was investigated in various specimens of the same type of crystal.
La sensibilité de cristaux à puits de dimensions différentes comme détecteurs du rayonnement quantique des radionuclides usuels

Les publications spécialisées contiennent des données établies au moyen de calculs théoriques, sur la sensibilité des cristaux d'iode de sodium ainsi que quelques données expérimentales sur la sensibilité des cristaux à puits en tant que détecteurs. Cependant, pour évaluer dans quelle mesure un cristal à puits se prête à la solution d'un problème de mesure déterminé, il ne suffit pas de connaître la sensibilité du détecteur. Pour savoir si un problème de mesure donné peut être résolu, il faut encore prendre en considération d'autres facteurs, tels que l'activité de fond, le temps disponible pour les mesures et le degré de précision voulu. L'auteur indique une méthode permettant de tenir compte de cet ensemble de facteurs.

Pour savoir si un problème peut être résolu, c'est la sensibilité qui constitue le facteur décisif; en revanche, pour l'étalonnage des sources radioactives, il est surtout utile de connaître le nombre d'impulsions par unité d'activité enregistrées par l'appareil considéré. Cette grandeur dépend essentiellement des caractéristiques de l'appareillage électronique associé au cristal. L'auteur indique, pour plusieurs ensembles, les sensibilités ainsi définies, telles qu'elles ont été déterminées par voie expérimentale pour quelques isotopes usuels. Dans le cadre d'un examen détaillé des erreurs, l'auteur rend compte de recherches sur les pertes au comptage relevées lors de mesures faites à l'aide de cristaux à puits. Il étudie également, pour différents exemplaires du même type de cristaux, la dispersion des valeurs pour une sensibilité déterminée.

Chuствительность различных размеров кристаллов при их бурении с помощью облучения квантовыми лучами обычных радиоизотопов

В литературе имеются теоретически обоснованные данные о чувствительности кристаллов натрия и иода, а также некоторые экспериментальные данные о чувствительности самого кристалла при бурении. Однако для определения целесообразности использования метода бурения кристалла с целью использования его при определенных измерениях чувствительность на детекторе является недостаточной. Если эта проблема будет решена, то лишь при учете дополнительных данных, таких как эффект порядка 0, может быть определено необходимое время измерения и достигнута требуемая точность. Ниже приводится метод определения чувствительности.

В то время, как чувствительность является главным фактором в решении рассматриваемой проблемы, для калибровки радиоактивных источников важную роль играет показатель числа импульсов на единицу активности. Этот показатель зависит, главным образом, от особых свойств электронного оборудования, используемого при бурении кристалла. Для различного комплекса приборов чувствительность аналогична при использовании различных изотопов. В целях определения погрешностей при установлении чувствительности проводилась проверка потери чувствительности при бурении. Затем исследуется полоса рассеивания чувствительности при различных образцах того же самого кристалла.

La sensibilidad de los cristales de cavidad de distintos tamaños a la radiación cuántica emitida por los radionúclidos corrientes

En la literatura especializada figuran datos teóricos acerca de la sensibilidad de detección de los cristales de NaI, así como algunos datos experimentales
sobre la sensibilidad de los cristales de cavidad. Ahora bien, con objeto de
evaluar la utilidad de un cristal de cavidad para determinado tipo de medición,
no basta conocer la sensibilidad del detector. Solamente si se toman en con-
sideración distintos factores, como el efecto cero del dispositivo, el tiempo
disponible para la medición y la precisión exigida, puede determinarse si el
problema de que se trata tiene solución. El autor propone un método que
permite tener en cuenta estos factores.

Para saber si un problema es soluble, es esencial conocer la sensibilidad,
et qno que para el calibrado de fuentes radiactivas es más importante conocer
e el número de impulsos que el dispositivo registra por unidad de actividad. Este
valor depende esencialmente de las características del dispositivo electrónico
asociado al cristal de cavidad. Para diversos tipos de aparatos, el autor indica
las sensibilidades así definidas y describe su determinación experimental en el
caso de algunos isótopos corrientes. Una discusión detallada de las fuentes de
error cierra el examen de las pérdidas experimentales durante el recuento con
ayuda de cristales de cavidad. Para una sensibilidad determinada, el autor
estudia además la dispersión de los valores observada con distintas muestras de
cristales del mismo tipo.

The most accurate method at present used in the determination of the
activity of radionuclide solutions is without doubt that of counting with the
4π counter. However one of the biggest disadvantages associated with this
method is the complicated preparation necessary before the start of the
measurements. Other methods are therefore used where the accuracy required
is not so important. One of these methods, which can be used when the decay
of the radionuclide to be measured is accompanied by quantum radiation, is
the activity measurement with the help of a scintillation counter and a well-
type crystal. It makes no difference whether this radiation is primary γ radiation,
or secondary radiation such as bremsstrahlung or characteristic radiation. In the
following articles the determination of the sensitivity of various well-type
crystals will be described, so that an estimation can be made as to which type
is most suitable for a particular series of measurements. The variables which
influence the sensitivity will also be discussed.

Naturally enough the sensitivity, which is given in \( \text{imp}_{\text{min}}/\mu\text{c} \), is not the
only variable which must be considered when estimating the suitability of a
particular well-type crystal for a particular measurement. The background
count of the measuring arrangement, the time of measurement and the accuracy
required also play a part. When

\[
E = \text{the sensitivity of the arrangement in } \frac{\text{imp}_{\text{min}}}{\mu\text{c}}
\]
\[
A = \text{the activity to be measured, in } \mu\text{c}
\]
\[
n_o = \text{the background count, in } \frac{\text{impulse}_{\text{min}}}{\mu\text{c}}
\]
\[
t = \text{time of measurement in minutes}
\]
\[
f = \text{the permissible relative statistical error}
\]

then the following expression holds

\[
t = \frac{E A + 2 n_o}{f^2 (EA)^2}
\]
This can be expressed in the form of a three dimensional diagram (1), as \( n \) is a constant for the measuring arrangement, and \( E \) is a constant for the measuring arrangement and a given nuclide (see figure 1).

![Figure 1](image)

**Figure 1**

Time of measurement-activity diagram for a well-type crystal Z11 by an extrapolated to zero charge depth for Co\(^{60}\) solution.

This seemingly complicated representation must always be used when comparing two detecting arrangements with regard to the respective lower threshold of measurement. A simpler specific number is desirable, even if it does not permit as general a deduction as that available from the time of

Measurements of the Tested Typs of Well-Typ Counters

![Figure 2](image)

**Figure 2**

Measurements and dates of the tested types of well-type crystals.
Measurement-Activity Diagram in figure 1. We can define such a specific number as the “statistically reliable threshold of measurement”. This is the activity which, in the given measuring arrangement, causes an impulse rate which in turn is the same as the 3σ threshold of the background distribution with a time of measurement of 1 minute. Through the above definition it is hoped to encourage a discussion about the most practical way of achieving a specific number for two measuring arrangements. The thresholds mentioned in the course of this talk are to be understood as defined above. Only the background and the sensitivity of the measuring arrangement play a part in the so defined specific number.

The sensitivity of different types of well-type crystals were therefore experimentally determined.

Investigations into the efficiency of scintillation counters with NaI crystals have been carried out by various people. For non-well-type crystals the efficiency for the events which fall in the photopeak has been calculated for various geometrical arrangements and various energies (2). However only one experimental investigation in the efficiency of well-type crystals is known to me (3). We will, therefore, discuss the factors which have an influence on the sensitivity of a well-counter arrangement.

1. Choice of the Applied Voltage or the Discriminator Threshold

On investigation of the characteristics of the scintillation-counter arrangement, with following integral discriminator, one finds no such ideal plateau with a low gradient as is obtained with a Geiger-Müller counter. Figure 3 shows some characteristics for various radionuclides.
The case of Ce\textsuperscript{144} is particularly interesting. Here the high-energy \( \beta \) component from the daughter product Pr\textsuperscript{144} is measured at first, and only by higher multiplier voltages are the impulses from the Ce\textsuperscript{144} \( \gamma \) radiation so big that they pass through the integral discriminator. For this reason one always obtains a curve with a relatively steep gradient, the form depending on the disintegration scheme. The determination of Sr\textsuperscript{90} + Y\textsuperscript{90} is carried out using bremsstrahlung, even though, as we will see later, a considerable number of impulses are caused by \( \beta \)-rays.

High-voltage supply units are now available which are so stabilized that the error due to the plateau gradient can be neglected, provided that the working voltage is carefully chosen.

2. Counting Losses

The decay-time of NaI scintillations and the resolving power of the scintillation counters are so short (about \( 0.25 \times 10^{-6} \) sec), that with the usual instruments the dead-time of the amplifiers and the counting circuits influence the result. The counting losses are dependent on the electronics which follow, and must be determined, for example using the Ba method. The measuring arrangement which we used in our experiments had a resolving power of approximately \( 10^{-6} \) sec for periodic impulses and the first counting losses occurred around \( 2.5 \times 10^{6} \) imp/min and were corrected for. Detailed investigations into the resolving power of scintillation counters and the following electronics have been published recently by GATTI and SVELTO \(^4\).

![Figure 4](image-url)

Changes in the sensitivity caused by the addition of layers of Aluminium for the well-type crystal Z11

Sensitivity in % with respect to the sensitivity without layers

<table>
<thead>
<tr>
<th></th>
<th>Co\textsuperscript{60}</th>
<th>Ce\textsuperscript{137}</th>
<th>Ce\textsuperscript{144} + Pr\textsuperscript{144}</th>
<th>Sr\textsuperscript{90} + Y\textsuperscript{90}</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thickness of layers</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>100%</td>
<td>100%</td>
<td>100%</td>
<td>100%</td>
</tr>
<tr>
<td>1</td>
<td>90%</td>
<td>90%</td>
<td>70%</td>
<td>50%</td>
</tr>
<tr>
<td>2</td>
<td>70%</td>
<td>70%</td>
<td>50%</td>
<td>30%</td>
</tr>
<tr>
<td>3</td>
<td>60%</td>
<td>60%</td>
<td>40%</td>
<td>20%</td>
</tr>
<tr>
<td>4</td>
<td>50%</td>
<td>50%</td>
<td>30%</td>
<td>10%</td>
</tr>
</tbody>
</table>

Figure 4
Dependence on wall thickness

380
3. Dependence on the Wall Thickness of the Container

The active solution is normally put into the well in a container, usually a glass ampoule. In figure 4 the sensitivity is plotted against the wall-thickness of the container. The diagram was obtained by putting Al cylinders of known thickness over the glass ampoule containing the radioactive solution. It is shown that the $\beta$ components of Co$^{60}$ (0.312 MeV) and Cs$^{137}$, (0.52 MeV) are screened out by the ampoule and the housing of the well crystal, and that the error for the $\gamma$ radiation of Cs$^{137}$, with 2.5 mm Al, is about 4%. The unavoidable variation in the wall thickness can therefore be disregarded for all $\gamma$ radiation which is not too soft. The expected error for a difference in thickness of 0.5 mm in dependence on the quantum energy is shown in Table I.

**TABLE I**

<table>
<thead>
<tr>
<th>Quantum energy</th>
<th>expected error</th>
</tr>
</thead>
<tbody>
<tr>
<td>20 keV</td>
<td>41 %</td>
</tr>
<tr>
<td>50 keV</td>
<td>5 %</td>
</tr>
<tr>
<td>100 keV</td>
<td>2 %</td>
</tr>
<tr>
<td>500 keV</td>
<td>1 %</td>
</tr>
<tr>
<td>1000 keV</td>
<td>0.8%</td>
</tr>
</tbody>
</table>

Ce$^{144}$ + Pr$^{144}$, and Sr$^{90}$ + Y$^{90}$ show another type of behaviour. Here part of the hard $\beta$ radiation passes through the container walls into the crystal, and therefore changes in the wall thickness cause appreciable changes in the sensitivity. Reproducible results for radionuclides with hard $\beta$ radiation can only be obtained by increasing the wall-thickness and for pure $\beta$ sources only the bremsstrahlung component is used.
4. Dependence on the Charge Depth

The dependence of the sensitivity on the charge depth was determined for a number of radionuclides. It has been found that a dependence on the volume of solution used, as is occasionally mentioned (3), can not be reproduced. Containers of varying diameter give varying charge depths. On the other hand with constant charge depth the sensitivity is nearly independent of the diameter of the solution, when the charge depth is less than 2/3 the well depth, and the changes in diameter are not too large.

Figure 5 shows the dependence on the charge depth. The change in the sensitivity increases with a decrease in energy. If we suppose that the charge depth can be fixed to within 1 mm, then the error lies below 0.5% at 600 keV, below 1% at approximately 100 keV. The effect due to the varying shape of the bottom of the container is even smaller, and can not be easily demonstrated. It is therefore to be recommended that the same sort of container be used throughout the experiments.

5. The Statistical Fluctuations of the Sensitivity of the Crystals

When the sensitivity for a number of different crystals of the same type is measured in the same arrangement and with the same voltage on the multiplier it is found to vary from crystal to crystal. The variation of the sensitivity over 10 crystals was measured, and the mean deviation was 2.4%. We were also able to show that the deviations in the sensitivity are caused by the varying luminescence yield of the crystals. If the crystals are investigated in an arrangement whereby the occurrences of the same energy loss in the crystal are turned into impulses of the same height, then the same sensitivity, for a particular radionuclide, is obtained at the output of the integral discriminator, for all the crystals, within the experimental accuracy. The "sensitive crystals" have therefore only a higher luminescence yield, and give rise to more impulses which lie above the discriminator threshold.

6. Accuracy of the Method of Measurement

In estimating the error of a method for the measurement of the activity of a solution of a radionuclide one must distinguish between three types of errors:

a) Errors, which occur during the preparation before the measurement. These errors can be kept very small for well-counter measurements as practically no preparation is required.

b) Errors, which occur in every measurement of radioactive substances, and which are caused by the statistics of the radioactive decay. These errors can be calculated from equation (1), by solving for f.

c) Errors, caused by the special measuring method when a well-type crystal is used. These are the uncertainty in the wall thickness of the container and the uncertainty in the charge depth. Both errors are dependent on the γ energy emitted by the radionuclides. As upper limit for this "special error" we can take the values given in Table II.
TABLE II
“Special error” in activity measurements a well-type counter

<table>
<thead>
<tr>
<th>Quantum energy</th>
<th>special error</th>
</tr>
</thead>
<tbody>
<tr>
<td>1000 keV</td>
<td>5.5%</td>
</tr>
<tr>
<td>50 keV</td>
<td>2.4%</td>
</tr>
<tr>
<td>100 keV</td>
<td>1.1%</td>
</tr>
<tr>
<td>500 keV</td>
<td>0.9%</td>
</tr>
</tbody>
</table>

From this we can see that the “special error” has, in most cases, the largest part in the total error, as the preparation error is very small and the statistical error mentioned under (b) can be neglected when the time of measurement is sufficiently long.

From Table II we can see that, in particular for hard γ radiation, the method using the well-type crystal for the determination of the activity of solutions of radionuclides is one which is astonishingly accurate, and has the advantage that it can be carried out quickly and without much trouble. In Table III the sensitivities for various crystals and radionuclides are shown.

TABLE III
Sensitivity of the well-type crystal Z 11 Nr. 256/226/356/312 measured with:
Scintillation counter FH 421, Nr. 1032, Voltage 1400 V
Amplifier FH 547, Threshold 50 mV
Counter: “FH, MHz = Einschub”
The sensitivity is corrected for zero charge depth.
Background: 400 imp/min

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Sensitivity</th>
<th>Efficiency</th>
<th>Statistically reliable threshold of measurement</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>imp/μc</td>
<td>%</td>
<td>μc</td>
</tr>
<tr>
<td>Co⁶⁰</td>
<td>$10.3 \times 10^8$</td>
<td>48 %</td>
<td>$0.6 \times 10^{-3}$</td>
</tr>
<tr>
<td>Au¹⁸⁸</td>
<td>$8.5 \times 10^8$</td>
<td>38 %</td>
<td>$0.8 \times 10^{-3}$</td>
</tr>
<tr>
<td>I¹³¹</td>
<td>$7.7 \times 10^8$</td>
<td>35 %</td>
<td>$0.8 \times 10^{-3}$</td>
</tr>
<tr>
<td>Cs¹³⁷</td>
<td>$6.1 \times 10^8$</td>
<td>27 %</td>
<td>$1.1 \times 10^{-3}$</td>
</tr>
<tr>
<td>Ce¹⁴⁴</td>
<td>$5.0 \times 10^8$</td>
<td>22 %</td>
<td>$1.4 \times 10^{-3}$</td>
</tr>
<tr>
<td>Sr⁹⁰</td>
<td>$1.2 \times 10^8$</td>
<td>5.2 %</td>
<td>$6.5 \times 10^{-3}$</td>
</tr>
<tr>
<td>F¹³⁷</td>
<td>$0.4 \times 10^8$</td>
<td>1.7 %</td>
<td>$24.0 \times 10^{-3}$</td>
</tr>
</tbody>
</table>

Sensitivity of the well-type crystal Z 20 Nr. 458/2251/2581
Same conditions
Background: 400 imp/min

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Sensitivity</th>
<th>Efficiency</th>
<th>Statistically reliable threshold of measurement</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>imp/μc</td>
<td>%</td>
<td>μc</td>
</tr>
<tr>
<td>Co⁶⁰</td>
<td>$14 \times 10^8$</td>
<td>62 %</td>
<td>$0.5 \times 10^{-3}$</td>
</tr>
<tr>
<td>Cs¹³⁷</td>
<td>$9 \times 10^8$</td>
<td>40 %</td>
<td>$0.9 \times 10^{-3}$</td>
</tr>
<tr>
<td>Ce¹⁴⁴</td>
<td>$6.3 \times 10^8$</td>
<td>28 %</td>
<td>$1.0 \times 10^{-3}$</td>
</tr>
<tr>
<td>Sr⁹⁰</td>
<td>$1.4 \times 10^8$</td>
<td>3.2 %</td>
<td>$5.0 \times 10^{-3}$</td>
</tr>
</tbody>
</table>

REFERENCES
DISCUSSION

Dr. Trott (United Kingdom):

I would like to make a few comments on Dr. Waechter's paper. As I understand it, he has been investigating how sensitivity varies with different types of well-crystal. We have been using well crystals for a considerable time and for various jobs and have recently been trying to decide what type of well crystal to use for measuring high-energy γ emitters which are natural-series nuclides. Now a point that came up here in discussion with the manufacturers (the Hershaw Company) when we were trying to get a 6'' × 5'' well crystal with, in our case, a rather deep well, was the problem of the choice of photomultiplier. At first they recommended that even with a 5'' diameter crystal we should use a 3'' photomultiplier with a special mounting of their design, because of the very much improved resolution that was obtained. Now I wonder if this new parameter — i.e., the fact that as you change the size of your crystal you are confronted with problems in photomultiplier sensitivity — has been considered by Dr. Waechter. Incidentally, in that connexion I believe the situation changes rather fast and EMI 5'' photomultipliers may now be quite suitable.

Furthermore, Dr. Waechter's method apparently does not require special preparation of sources but can be used to measure strontium by bremsstrahlung. This would seem likely to run into difficulties as regards reproducibility, because changes in size, form etc. of the source will certainly affect the bremsstrahlung emitted.

My final point is that, although this seems sure to be a method which can give good precision when calibrated, very careful control by the use of carefully chosen reference sources is necessary. For example, if you are measuring Cr$^{51}$ it is no good setting up your system using Co$^{60}$ because it may well give constant sensitivity there, but would not apply for chromium. We, for example, use the bremsstrahlung from the strontium source to control the system.

Dr. Waechter (Federal Republic of Germany) (Translated from French):

I may perhaps begin with the use of bremsstrahlung. We used very low-content solutions and so I do not think that the nature of the substance greatly affects the yield of the bremsstrahlung. I agree with Mr. Trott that the substance greatly influences the output of bremsstrahlung where widely different substances are involved. Consequently only sources of virtually identical substances can be compared.

As regards the size of the well crystals, we have used only sources with γ radiation of just a few MeV. The hardest γ radiation we have used was that of Co$^{60}$, though not in the experiments described, and there were no difficulties. Crystal size is the main factor in measurements of very hard radiations, but as crystal size increases, the yield of the luminescence becomes more uneven. This may lead to difficulties where very hard radiations are measured.
III.

NEUTRON SOURCE CALIBRATION

(SESSION 5, 16 OCTOBER 1959)
III. 1. LOW LEVEL STANDARDIZATION PROGRAMME AT RISO

J. THOMAS, P. SKJERK CHRISTENSEN, BANT FASTRUP, J. OLSEN AND P. VILLEMOES

Abstract

In order to obtain a calibrated neutron flux several methods have been employed. A comparison is made of the results obtained by the different methods.

Programme d'étalonnage de sources de neutrons peu intenses à Risö

Plusieurs méthodes ont été employées pour obtenir un flux de neutrons calibré. On compare les résultats obtenus par les différentes méthodes.

Программа стандартизации низких уровней радиации в Ризо

Для получения калиброванного нейтронного потока разработано несколько методов. Проведено сравнение результатов, полученных различными способами.

Programa de calibración de fuentes neutrónicas poco intensas desarrollado en Risö

El autor ha empleado varios métodos para obtener un flujo neutrónico calibrado. Compara los resultados conseguides con los distintos métodos.

Introduction

The measurements are carried out in the Danish reactor DR 1 at Risö (5). DR 1 is of the water boiler type, Atomic International Model L-55, operating at a power level of 2000 W. The reactor went critical in August 1957. It consists of a 32 cm diameter and 1.75 mm wall spherical core containing about 5 kg uranium enriched to 20% in U\textsuperscript{235}. The reactor is surrounded by a 1.3 meter high, 1.5 meter diameter cylinder of graphite which acts as reflector. Along a diameter of the spherical core is a 2.5 cm inner diameter tube allowing access to the center of the sphere. In addition, it is possible to irradiate samples in eight stringer holes in different horizontal parallel planes in the graphite. The reactor is cooled by a coil immersed in the core solution.

Activation Method

We have adopted WESTCOTT's convention (3) on reactor spectra, i. e.

\[ \mathcal{O}(E) = F_M \left\{ \frac{E}{(E_T)^2} e^{-E/E_T} + \beta \frac{\Delta}{E} \right\} \]

where

\[ E_T = k \ T \text{ and } \Delta = \begin{cases} 0 & \text{for } E < \mu kT \\ 1 & \text{for } E > \mu kT \end{cases} \]

The factor $\beta$ is conveniently calculated from measured cadmium ratios (3). By the thermal flux we understand $n^M v_0$, where $n^M$ is the integral neutron density in the Maxwell distribution. The thermal reaction rate is then calculated as $(n^M \cdot v_0) \sigma_0 g$, where $g$ is a factor introduced by Westcott, $\sigma_0$ is the activation cross-section at 2200 m/sec, and $g$ is 1 for an $1/v$ absorber in the thermal region.
For non-1/v absorbers $g$ is a function of neutron temperature $T$, so we have to measure or calculate $T$. $(n^Mv_0)$ is determined from the difference of reaction rates of a thin gold foil, when irradiated bare and cadmium covered.

$$R - R_{cd} = \int_0^{\infty} \sigma(E) \phi(E) \, dE - \int_{E_{cd}}^{\infty} \sigma(E) \phi(E) \, dE$$

The flux expression is transformed to:

$$R - R_{cd} = F_M \sqrt{\frac{\pi T_0}{4T}} \sigma_0 + \beta F_M \int_{E_{cd}}^{E_{fcd}} \sigma(E) \, dE$$

where $F_M = \left( \frac{\pi T_0}{4T} \right)^{1/2} n^M v_0$

$E_{cd} = 0.4 \text{ eV}$, $\mu = 2.81$ (light water moderator), but the values are not critical for small $\beta$.

In order to determine a thermal neutron flux it is necessary to measure the flux perturbation and self-shielding of the foils. This we have done for gold in graphite by measuring the activity in foils ranging from about $0.3 \text{ mg/cm}^2$ to $100 \text{ mg/cm}^2$. The foils were $1 \text{ cm}^2$, some punched out of sheets and some evaporated on thin films. The foils were irradiated bare and cadmium covered in the same position, and counted in a $4\pi\beta$ counter as well as in a $\gamma$ counter. To determine the activation from the thermal neutron flux we calculate the difference between the activity from the bare foils and the cadmium covered foils after corrections for the absorption of epithermal neutrons in cadmium. Here we use values of $F_{cd}$ given by MARTIN. During the measurement the foils were placed on a thin film made of formvar (polyvinyl formal). The thickness of the films was about $25 \mu\text{g/cm}^2$ with a $5 \mu\text{g/cm}^2$ conducting film of aluminium. The only correction for the $4\pi\beta$ counter is the self-absorption for the $\beta$ particles in the foils. By comparison of the counting rates in the $4\pi\beta$ and the $\gamma$ counters we directly have the combined effects of flux perturbation and self-shielding and in addition the self-absorption of $\beta$ particles in gold, all from thermal activation. By using the above corrections the thermal neutron flux can be determined with an estimated error of $5\%$.

In addition to the $4\pi\beta$ proportional counter which consists of two hemispherical chambers with ring-shaped anodes, a $4\pi\beta-\gamma$ coincidence unit similar in principles to that used by CAMPION is now in current use.

**Calorimetric method**

Using the reactor as a calorimeter (neglecting the neutrinos and parts of the $\gamma$-rays) the flux can be determined from the heat generated by the reactions taking place in the core.

The heat output is measured by the temperature rise and the flow-rate of the cooling water when the reactor is stationary.

The temperature rise is measured by a pair of resistance elements immersed in the inlet and outlet water-lines. The temperature-dependent resistances are measured in a Wheatstone bridge. The temperature rise is approximately 6 degrees centigrade at 2 kW when the flow is 300 litres per hour.

The flow-rate is measured by a venturi-type flow-meter calibrated volumetrically.

The error in measuring the heat output is estimated to $5-8\%$ including
the uncertainty in power transport between the core and the system which is recombining the produced radiolytic gas.

From the power we can calculate the maximum thermal flux \( n^{M \nu_0} \) as follows:

The power \( P \) is given by

\[
P = R_f \ W
\]

where

- \( R_f \): the energy released per fission
- \( W \): the total fission rate

Now

\[
R_f = n^{M \nu_0} (1 + F) \Sigma_{f_0} \ C
\]

where

- \( n^{\nu_0} \): the thermal flux in the center of the core, in Westcott's notation
- \( F \): the fraction of fissions occurring above \( \mu \)kT
- \( \Sigma_{f_0} \): the macroscopic fission cross-section at 2200 m/sec
- \( C \): a geometry factor

\( \Sigma_{f_0} \) is calculated from the composition of the core.

Now

\[
n^M (r) \nu_0 = n^{M \nu_0} \ \frac{\sin(kr)}{(kr)}
\]

where \( r \) is the distance from the core-center. From this \( C \) is defined as

\[
C \ n^{M \nu_0} = \int n^M (r) \nu_0 \ dV = n^{M \nu_0} \int \frac{\sin(kr)}{kr} \ dV
\]

where the integration is over the volume of the solution.

The factor \( k \) is determined as follows:

The actual thermal flux distribution is found by irradiating bare and cadmium covered indium foils in the control exposure tube. The foils were 100 mg/cm\(^2\) thick. The activity was measured by \( \gamma \) counting. The difference between the activities is a quantity proportional to the thermal flux, and the obtained flux curve is fitted to \( \frac{\sin(kr)}{kr} \) in order to determine \( k \). This can be done with an error of some 5\%.

Using the above equations with appropriate constants \((6) (3)\) the power is given in terms of \( n^{M \nu_0} \) as:

\[
P = 3.72 \times 10^{-8} \ n^{M \nu_0} \ \text{watts}
\]

where \( n^{M \nu_0} \) is the thermal flux in the center of the core. The error is approximately 10\%.

Comparison

Using the linear neutron-level recorder as reference the thermal neutron flux calculated by the two before-mentioned methods is compared.

A difference of 2\% is found.

Source strength

The source strength of a 100 mc Ra-Be source is measured by a student as an examination problem. The work was carried out with the use of manganese-
sulphate as an absorber and measurements resulted with an estimated error of 3%. The report will soon be finished.

\( \gamma - \gamma \) coincidence

In order to get the full use of a \( \gamma - \gamma \) coincidence equipment we have been considering the total number of corrections which have to be used when the efficiencies of the detectors are not small.

According to BELL (2) an arrangement with an anti-Compton shield will strongly reduce the effect of scattering of radiation from one crystal to the other. We, therefore, neglect this effect and have then that only real coincidences will be counted in the coincidence circuit.

We now assume that the source emits two gammas \( \gamma_1 \) and \( \gamma_2 \) with the spectra \( f_{11}(E) \) and \( f_{21}(E) \) respectively where the spectra in this case means the electronics signal spectra in the first detector that the \( \gamma \) would produce if it was emitted as a single line i.e. not in time coincidence with the other line.

Let now \( W_{11} \) denote the total efficiency of the first counter for the first \( \gamma \) line (as single line) and \( W_{21} \) the total effectivity of the first counter for the second \( \gamma \) line (as single line).

If we introduce a discriminator-setting \( d \), we only count a part of the spectrum and we then get a discriminating effectivity defined by

\[
\epsilon_{11}(d) = \frac{\int d_1(E, d) \cdot f_{11}(E) \, dE}{\int f_{11}(E) \, dE}
\]

and

\[
\epsilon_{21}(d) = \frac{\int d_1(E, d) \cdot f_{21}(E) \, dE}{\int f_{21}(E) \, dE}
\]

where \( d_1(E, d) \) is the transmission function of the discriminator in the first detector system.

If the two \( \gamma \) enter the first crystal simultaneously we get a “sum” spectrum \( f_{s1}(E) \) given by

\[
f_{s1}(E) = \int_0^E f_{11}(t) \cdot f_{21}(E-t) \, dt
\]

and the discriminator effectivity for this spectra is

\[
E_{s1}(d) = \frac{\int dE \cdot d_1(E, d) \cdot \int_0^E f_{11}(t) \cdot f_{21}(E-t) \, dt}{\int dE \int_0^E f_{11}(t) \cdot f_{21}(E-t) \, dt}
\]

We now get for the counting rate in the first counter
\[ N_1 = Q \left\{ (W_{11} - W_{11} W_{21}) \varepsilon_{11} (d) + (W_{21} - W_{11} W_{21}) \varepsilon_{21} (d) + W_{11} W_{21} \varepsilon_{51} (d) \right\} \]

\[ = Q \left\{ W_{11} \varepsilon_{11} (d) + W_{21} \varepsilon_{21} (d) + W_{11} W_{21} \left[ \varepsilon_{51} (d) - \varepsilon_{11} (d) - \varepsilon_{21} (d) \right] \right\} \]

In the same way we get for the second counter

\[ N_2 = Q \left\{ W_{12} \varepsilon_{12} (d) + W_{22} \varepsilon_{22} (d) + W_{21} W_{22} \left[ \varepsilon_{52} (d) - \varepsilon_{12} (d) - \varepsilon_{22} (d) \right] \right\} \]

In the coincidence channel we get

\[ N_c = Q \left\{ W_{11} \varepsilon_{11} (d) W_{22} \varepsilon_{22} (d) + W_{12} \varepsilon_{12} (d) W_{21} \varepsilon_{21} (d) \right\} \]

where

\[ Q \] is the source strength.

An investigation of the \( \gamma-\gamma \) coincidence following the line shown here is in progress and we hope to be able to give a practical procedure for handling these corrections.

List of symbols

- \( d \) = discriminator setting
- \( d_j (E, d) \) = transmission function of the discriminator in detector \( j \)
- \( E \) = neutron or \( \gamma \) energy
- \( E_{\text{cd}} \) = cadmium cut-off energy
- \( \varepsilon \) = discriminator effectivity
- \( f_{ij} (E) \) = electronics signal spectra of \( \gamma \) (as single line) in detector \( j \)
- \( g \) = quantity concerning the departure in the cross-sections from the \( 1/\nu \) law. It is defined by \( R^M = \sigma_0 g N^M N \)
- \( k \) = Boltzmann's constant
- \( \mu \) = adjustable parameter (in this paper \( \mu = 2.81 \))
- \( N \) = counting rate
- \( n^M \) = integrated thermal neutron density
- \( Q \) = source strength
- \( R \) = total reaction rate
- \( R_{\text{cd}} \) = reaction rate due to the resonance neutrons \( (E > E_{\text{cd}}) \)
- \( R^M \) = reaction rate due to the thermal neutrons
- \( R_f \) = total fission rate
- \( \sigma_0 \) = activation cross-section at neutron velocity 2200 m/sec
- \( T \) = neutron temperature
- \( T_0 = 293.6 \) °K
- \( N_0 = 2200 \) m/sec
- \( W \) = energy released per fission in \( U^{235} \)
- \( W_{ij} \) = total effectivity [according to BELL (2)] of detector \( j \) for \( \gamma_i \) (discriminator zero).

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III. 2. ABSOLUTE $\beta$ COUNTING AND ITS APPLICATION FOR THE STANDARDIZATION OF RADIOACTIVE NEUTRON SOURCES

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Abstract

The emission rate of the Canadian neutron standard, a Ra-Be ($\alpha$, n) source, has been determined by two methods. Both involve the standardization of radioactive nuclides.

By the first method, the neutrons were moderated in a manganese sulphate solution surrounding the source. A calculable number of thermal neutrons is captured in the manganese of the bath and produces radioactive Mn$^{56}$. From its resulting activity the neutron emission rate is found. High specific activity Mn$^{56}$ solution, prepared by using the Szilard-Chalmers process, was standardized by 4 $\pi$ $\beta$ counting (proportional) and compared with the activated solution of the bath.

The second method which does not seem to have been used previously, makes use of the F$^{19}$ ($\alpha$, n) Na$^{22}$ reaction in a specially prepared Po-F source. Since fluorine is monoisotopic the neutron output can be calculated from the yield of the resulting radioactive Na$^{22}$ because for each neutron a Na$^{22}$ nucleus is produced. Na$^{22}$ is a positron emitter and discrimination against $\gamma$-rays from Po$^{210}$ and from the reaction could be achieved by detecting the two annihilation quanta in coincidence. Comparison was made with a standardized Na$^{22}$ source. Standardization was carried out using the 4 $\pi$ $\beta$-$\gamma$ coincidence technique; details on its application to Na$^{22}$ will be described.
Абсолютный счет $\beta$ частиц и его применение для стандартизации радиоактивных нейтронных источников

Интенсивность испускания канадского нейтронного стандартного источника Ra-Be ($\alpha$, $n$) определяется двумя способами. Оба способа связаны со стандартизацией радиоактивных изотопов.

С помощью первого метода нейтроны замедляются в растворе сульфата марганца, который окружает источник. Подсчитываемое количество тепловых нейтронов захватывается в марганце раствора, в результате чего образуется радиоактивный $\text{Mn}^{56}$. Интенсивность нейтронной эмиссии определяется по его конечной активности. Радиоактивный раствор $\text{Mn}^{56}$ особой активности, изготовленный путем использования процесса Сцилард-Чальмерса, был стандартизован 4 $\pi$ $\beta$ счетчиком (пропорциональным) и сравнен с активированным раствором ванны.

Второй метод, который, видимо, не употреблялся ранее, заключается в использовании реакции $\text{F}^{19}$($\alpha$, $n$) $\text{Na}^{22}$ в специально подготовленном источнике полоний-фтор. Поскольку фтор является одноизотопным, выход нейтронов может быть подсчитан из выхода результирующего $\text{Na}^{22}$, так как на каждый нейтрон производится ядро $\text{Na}^{22}$. $\text{Na}^{22}$ испускает позитроны, и распознавание на фоне гамма-лучей, исходящих из $\text{Po}^{210}$ и получающихся в результате реакции, может быть достигнуто подсчетом совпадений от двух аннигиляционных квартов. Проведено сопоставление со стандартизованным источником $\text{Na}^{22}$. Проводится стандартизация путем использования метода 4 $\pi$ $\beta$-г счета совпадений и будут описаны подробности применения его к $\text{Na}^{22}$.

Recuento $\beta$ absoluto y su aplicación a la calibración de fuentes emisoras de neutrones

El autor ha determinado por dos métodos diferentes la intensidad del patrón neutrónico canadiense, constituido por una fuente de Ra-Be ($\alpha$, $n$). Ambos métodos exigen la calibración de núcleos radiactivos.

El primero consiste en moderar los neutrones en una solución de sulfato de manganeso colocada alrededor de la fuente. El manganeso de la solución captura un número calculable de neutrones térmicos, formándose $\text{Mn}^{56}$ radiactivo, cuya actividad permite evaluar el flujo neutrónico. La solución de $\text{Mn}^{56}$ de elevada actividad específica, que se prepara utilizando el proceso Szilard-Chalmers, se normaliza por recuento $\beta$ $4 \pi$ (proporcional) y se compara con la solución activada mencionada en primer término.

Para la segunda técnica, que al parecer no se ha utilizado anteriormente, se emplea la reacción $\text{F}^{19}$($\alpha$, $n$) $\text{Na}^{22}$ producida por una fuente de Po-F especialmente preparada. Como el flúor es monoisotópico, el flujo neutrónico puede calcularse sobre la base del rendimiento de $\text{Na}^{22}$ radiactivo, ya que por cada neutrón se forma un núcleo de $\text{Na}^{22}$. Los rayos $\gamma$ emitidos por el $\text{Na}^{22}$, emisor de positrones, pueden distinguirse de los que proceden del $\text{Po}^{210}$ y de la reacción, respectivamente, detectando por coincidencias los dos cuantos de aniquilación. Como referencia, el autor utilizó una fuente de $\text{Na}^{22}$ calibrada. Ejecutó la calibración mediante la técnica de coincidencias $\beta$-$\gamma$ $4 \pi$. En la memoria figuran detalles de la aplicación de este método al $\text{Na}^{22}$.

394
In the field of nuclear physics and radiation dosimetry the use of calibrated neutron sources is often necessary. Since neutrons are non-ionizing absolute standardization of such sources can not be carried out by a simple counting method. This is particularly so because source neutrons cover a wide energy range and accurate information on the energy distribution is rather scanty.

A great number of indirect methods however are available for calibration; most of these finally involve the determination of the disintegration rate of radioactive nuclides. For this reason it is interesting to mention neutron standardization at this Symposium. Anyone who has routine equipment for radionuclide standardization also has the basic tools for neutron standardization at his disposal.

The neutron standard of the National Research Council is a 200 mg Ra-Be source, using the Be9 (α, n) C12 reaction. The advantage of this source type is its small physical size. The main disadvantage is that its neutron emission rate is not constant. For a new source the rate increases by more than ½% of one percent per year due to the growth of Po210, an α-emitting decay product of the radium. Some people also claim that the radium-beryllium mixture may physically change over the years but there is no definite proof of this. In most sources the mixture is in the form of a pressed pellet which makes demixing extremely unlikely. Another possible choice for a neutron source is the plutonium-α-beryllium source which however has to be physically large to obtain a suitable neutron output. The same applies to photonutron sources which have the additional disadvantages that the neutron energy is much lower than that from Be (α, n). Most National Laboratories use Ra-Be (α, n) sources and a comparison with a photonutron source is complicated and requires additional corrections. All these considerations led to the choice of a Ra-Be (α, n) source as the NRC standard. Fortunately a source was available which was already 10 years old and the increase in neutron emission is now below 0.3% per year.

For absolute standardization two quite different methods were used; both however make use of absolute counting. In the first method the neutrons from the source are slowed down in a manganese solution and captured in the manganese. The resulting Mn56 activity is then determined. In the second method an auxiliary Po-α-F source is used. In the F19 (α, n) reaction for each neutron a radioactive Na22 nucleus is produced. Absolute counting of the resulting Na22 gives the neutron emission.

The first method is basically simple and has been used, with variations, several times. Here, an improved method for the manganese standardization is used. The neutron standardization is carried out as follows: The neutron source is placed into the center of a tank, filled with manganese sulphate solution. The tank is 90 cm in diameter, big enough so that only a few neutrons can escape. When the Mn56 activity has built up to equilibrium the number of manganese atoms disintegrating per second is equal to the number of neutrons captured by the manganese. Not all neutrons are captured by Mn and the neutron emission rate of the source is

\[ Q = Q_{Mn} \frac{N_{Mn} \sigma_{Mn} + N_S \sigma_S + N_H \sigma_H}{N_{Mn} \sigma_{Mn}} \]

where the \( \sigma \)'s are the corresponding absorption cross-sections for thermal neutrons (S for sulphur and H for the hydrogen in the solution). For the concentration used (near saturation), half of the neutrons were captured in the hydrogen.
After the source is removed the solution is stirred to distribute the activity evenly. The absolute Mn$^{56}$ activity then has to be determined. However the specific activity is far too low for a direct determination with a 4πβ counter. Only a relative measurement of the γ activity can be made by immersing a multicellular (for increase of γ sensitivity) Geiger-Müller counter.

An auxiliary experiment therefore had to be carried out to obtain an absolute interpretation of the relative measurement. A small quantity of Mn solution, about 1 liter, was irradiated with a 1 gram Ra-Be neutron source and the activity produced measured absolutely. This standardized solution can then be mixed into the inactive bath to obtain a calibration of the bath-multicellular counter assembly. However even this solution produces deposits on the 4π counter film too thick to be really suitable for β counting. Therefore a Szilard-Chalmers process on one litre of Ca(MnO$_2$)$_2$ solution was used. After irradiation the solution only needs to be filtered and the remaining few milligrams of MnO$_2$ contain the bulk of the Mn$^{56}$ activity. The specific activity increased by a factor of 5000. The MnO$_2$ was taken up with H$_2$SO$_4$ and, after evaporation of the acid, 50 ml of stock solution prepared by dissolving the highly active MnSO$_4$ in H$_2$O. The greater part of this was mixed into the tank to be counted with the multicellular counter.

From the remainder, counting sources were prepared with 50 λ aliquots and deposited on gold-plated VYNS films. Insulin was used as a spreading agent. Most of the sources were rather poor in that they did not dry evenly and

![Figure 1](image.png)

γ scintillation spectra of a Po-F (α, n) source, 12 months old. The lower spectrum is in coincidence with the neutrons, giving information on excited states of Na$^{22}$ (6).
gave unsatisfactory counting plateaux. Apparently this is due to the fact that MnSO₄ is hygroscopic. Reproducible results were finally obtained by using a method where the source is dried in an NH₃ atmosphere under infra-red light. This produced an even and very finely grained, brown deposit of MnO₂. The counting was carried out with the type of 4π proportional flow counter described by Dr. Campion in a previous paper. Mn⁵⁶ emits rather energetic β radiation; the end point energy of the least energetic branch is 0.7 MeV. With an average source thickness of about 50 μg/cm² no correction for self-absorption was applied; this was estimated to be well below 1%.

![Figure 2](image)

The result of several bath calibrations was that 3960 Mn⁵⁶ disintegrations are needed for one count in the multicellular counter. Since there is no convenient way to increase the efficiency of the relative counting system appreciably, any bath method is limited to neutron sources of rather high neutron emission; the minimum which can be used is about 10⁵ neutron/sec.

A value of the neutron emission from the source can now be obtained after a small correction is applied for neutrons escaping from the tank and for capture of fast neutrons in the bath. As an average over several measurements a value of 3.22 × 10⁶ neutrons/sec. was obtained with a total error of ±1.5%.

The F¹⁹ (α, n) Na²² reaction in a polonium-fluorine source is used in the second method. The Na²² decays by positron emission and electron capture with

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* Paper I. 1.
a half-life of 2.6 years. From the growth of its activity within the source the 
neutron emission can be calculated. The problem is to determine the absolute 
Na\textsuperscript{22} activity without opening the source. The calibration therefore has to be 
carried out by a \( \gamma \) measurement. Since there is concurrent \( \gamma \) emission from the 
polonium in the source and the nuclear reaction itself (figure 1) the positron 
anihilation radiation from Na\textsuperscript{22} was measured only. Since the annihilation 
quanta are emitted from the source simultaneously and in opposite directions 
they can very conveniently be detected in a coincidence arrangement. Figure 2 
shows how two scintillation counters are used for the coincidence measurement. 
Comparison was made with a Na\textsuperscript{22} standard contained in a capsule identical 
with that of the neutron source. Background effects could easily be eliminated 
by subtracting the coincidence rate obtained at a 90\(^\circ\) setting from that at the 
normal 180\(^\circ\) setting. This takes care of accidentals and coincidences without 
directional correlation.

The Na\textsuperscript{22} standard was obtained by \( 4 \pi \beta \) counting of samples from a 
Na\textsuperscript{22} solution of high specific activity. Samples were measured in the same 
proportional flow counter which was mentioned before. Because the decay of 
Na\textsuperscript{22} is accompanied by a 1.3 MeV \( \gamma \)-ray it can also be conveniently measured 
in a \( 4 \pi \beta-\gamma \) coincidence counter. Since the apparatus shown in Fig. 5\(^*\) was not 
completed at this time Dr. Campion of Atomic Energy of Canada Limited 
measured the self-absorption for positrons in his counter. It was found that

\[ (2.5 \pm 0.5) \% \text{ of the positrons were lost by self-absorption.} \]

To obtain the Na\textsuperscript{22} disintegration rate a further correction for the electron capture has to 
be taken into account, the theoretical value of 10.2\% was used which is 
supported by recent experimental determinations.

Figure 3 shows the growth of the Na\textsuperscript{22} activity in the source. The theoretical 
growth curve is fitted to the experimental points. The Na\textsuperscript{22} activity

\* Paper I. 1.
reaches a maximum after 500 days. The decay curve for Po$^{210}$ is also shown (T = 138 days), and obviously the neutron emission from the source decreases in the same way.

From the height of the growth curve the neutron emission from the source can be calculated. Because of its short life a Po-F source cannot conveniently serve as a standard and therefore had to be compared to the NRC Ra-Be standard. The spectra of these two sources are very different; the maximum energy for Ra-$\alpha$-Be is 11 MeV and for Po-$\alpha$-F 3 MeV (Figure 4). The inter-

![Neutron spectra from Po-F ($\alpha$, n) source and from Be ($\alpha$, n) source.](image)

Figure 4

Neutron spectra from Po-F ($\alpha$, n) source and from Be ($\alpha$, n) source (e.g., reference 1).

comparison can not be carried out with a paraffin-surrounded BF$_3$ counter which is used here routinely when the neutron spectra are similar. For comparison an energy independent detector is needed and the MnSO$_4$ bath has been used where all neutrons are thermalized before capture. In this case only dip-counting and no absolute manganese activity determination is necessary. For the NRC neutron standard a value of $3.16 \times 10^8$ neutrons/sec was found with an error of 3%. Both methods are therefore in close agreement. This second standardization method has the advantage that no cross-section values are needed but the error is greater, mainly because the Na$^{22}$ standardization could not be carried out in a straightforward manner. The source which originally contained 1.5 curies Po$^{210}$ has now decayed to a low enough level so that it is planned to open the source and to separate the sodium for a direct Na$^{22}$ determination on the 4$\pi$$\beta$ counter. Experiments also were carried out with separate $\alpha$ sources placed close to fluorine-containing crystals. But with $\alpha$ sources of a strength which can safely be used the neutron intensities were too low for their accurate measurement.

It should be emphasized again that, generally, the methods of absolute counting are very important for neutron flux determinations and any improvement there would be very worth while. An important application of absolute counting in neutron measurement is, for instance, the absolute determination of foil activities after such foils have been activated with fast or
slow neutrons. Another application is the absolute counting of charged particles which are products of neutron-producing nuclear reactions taking place on the target of an accelerator. This is the associated particle method, used to obtain a known neutron flux. The most common reaction is $^7\text{Li}(\text{D}, \text{n}) \text{He}_4$ which gives 14 MeV neutrons. Absolute counting of the $\alpha$ particles determines the neutron emission.

Just as in absolute counting the accuracy of the calibration of radioactive neutron sources has increased greatly during the last few years. Five years ago, differences of calibrations between various laboratories of up to 10% were not unusual. Today these differences have been reduced to about 4%. Figure 5 shows the results of intercomparisons LARSSON (2) has carried out in Sweden in 1958. According to this graph the relative strengths of nearly all sources deviate less than $\pm 2\%$ from the mean. This year the Canadian neutron standard was sent to various European laboratories. Not all results are available yet but it appears that the relative source strength of the Canadian standard would lie about 1% above the average shown in figure 5.

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DISCUSSION

Dr. B. W. Robinson (United Kingdom):
I should like to refer to Mr. Geiger's paper. As I mentioned in my paper on Wednesday, at the National Physics Laboratory, we have been making measurements on neutron comparisons and absolute measurement of sources by what is, I think, exactly the method which he described with the manganese tank. But I just wanted to check that I was correct in thinking that Mr. Geiger's comparisons referred to radium-α-beryllium sources and not to photoneutron sources. Most of the work we have done has been on the detachable γ-ray source. I should also like to thank Mr. Geiger particularly for his last slide, which indicates that the NPL result is of infinite accuracy.

Dr. Geiger (Canada):
I am glad you liked the last slide. Only the NBS source was a photoneutron source. We have made a very careful comparison in Ottawa and taken into account the last improvements of the corrections which are necessary for such a comparison.

Dr. Allen (United Kingdom):
Has Mr. Geiger found a difference between apparent half-life and the accepted Po\(^{210}\) half-life in his polonium-fluorine sources?

Dr. Geiger:
No. We have checked the decrease in the neutron emission rate very carefully over two years, and within the statistical errors it follows the published half-life of polonium; I believe this is a good sign that the source was physically stable. I should perhaps add that this was a polonium-calcium fluoride source, since you cannot just mix polonium and fluorine in a metal container. This source was made by Mount Laboratories.

Dr. Curran (United Kingdom):
I should like to ask Mr. Geiger if Cf\(^{252}\) as a standard neutron source has been examined at all in his inter-comparison work. The AWRE's studies on \(n_u\) determination — i.e., the average number of neutrons per fission for U\(^{235}\) — were largely based on such a source and seem to give rise to some doubt about the more or less accepted value of \(n_u\) as found from standard neutron sources, as they yield a lower \(n_u\) value than that hitherto adopted in most reactor studies. Incidentally, this shows how important such sources are.

Dr. Geiger:
I believe that spontaneous fission sources will one day make very good neutron standards. I did not have access to such sources, but I should be very happy to measure one in our laboratory. If you can help us, I should be glad.

Dr. Curran:
The discrepancies that we think we are finding in \(n_u\) are of the order of 6%. This is an extremely important matter, of course, from the reactor point of view, and we shall certainly try to bring these sources, if we can, into line with the sources described this morning and should be very glad to see accuracies of better than 1% achieved.

Dr. Geiger:
I do not know if we shall ever achieve accuracies of less than 1% on neutron-emission-rate measurements; they are extremely involved and not quite as straightforward as the measurements discussed here, like \(4\pi\) counting, etc. 26
Professor P. Huber (Switzerland):

Mr. Geiger, how accurate are the fluorine-polonium neutron source measurements?

Dr. Geiger:

The accuracy with this measurement was ± 3%, and ± 1.5% with the manganese-sulphate method. The fluorine-polonium method is not as good as the latter but is very different, which was why I thought it worth while to try it.

Dr. Trott (United Kingdom):

Dr. Geiger mentioned, in discussing the manganese tank measurements, that the activity obtained was too low for $4\pi$ counter measurement. In view of the desirability, on the grounds of the simplicity (though perhaps we should not say that after the last few days) of the $4\pi$ counting method, I wonder whether he could give us some indication of what he meant by too low, since using the low-level $4\pi$ counters, such as the ones we mentioned yesterday, one can go down, I should think, reasonably to better than 10% of normal level, perhaps even below that. I wondered if he could just say what kind of activity he would like to be able to measure. We are interested in using low-level $4\beta$ counters in reactor, high-energy and activation analysis studies.

Dr. Geiger:

We have usually used about 400 or 500 litres or even more of the solution and this, of course, gives you a very low specific activity and the solution is saturated with manganese. All that can be done is to make a $\gamma$ measurement, and if you want to have a high or just a relatively high detecting efficiency, you have to put very many $\gamma$ detectors into the tank.

Dr. Trott:

Is it not a reasonable proposition to separate the manganese precipitated in some straightforward ways? It is too big a volume to handle at all?

Dr. Geiger:

Well, I think people in some laboratory in Great Britain have done this by building a machine where the potassium permanganate solution was circulated through filters, a Szilard-Chalmers process being used, but to do this with the required reproducibility of 1% is very difficult and I doubt whether a Szilard-Chalmers process would give a reproducibility of such a high order.

Professor Aten (Netherlands):

We recently calibrated a photoneutron source by dividing a manganese bath into two parts — a relatively small inner part containing most of the activity and filled with manganese sulphate solution, and an outer part filled with potassium permanganate solution. The outer part, in which the smaller part of the activity occurs but which is larger than the inner part, can then be measured by means of the Szillard-Chalmers process, and the inner part, containing the smaller but most active part of the liquid, can be measured directly after some of the manganese has been separated.

Dr. Geiger:

I believe that this is a very good idea, but what I am personally looking for is some method by which you can simplify the whole question of neutron measurements. I do not know of a way yet, but I believe that in some years we may have a better method.
IV.

HIGH INTENSITY SOURCE MEASUREMENTS

(SESSION 5, 16 OCTOBER 1959)
IV. 1. THE CONCEPT OF "GRAMME-EQUIVALENT-OF-RADIUM" FOR NON-POINT ISOTOPIC γ-RAY SOURCES

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Abstract

It is very useful to use the concept of "gramme-equivalent-of-radium" (gRa) in industrial radiography and in teletherapy to express the activity of an isotopic γ-ray source. This concept, while giving an idea of the activity of the source, permits at the same time to determine dose-rates in r/h to be expected at any distance from the source.

There exists a difference of opinion as to the concept itself of the "gramme-equivalent-of-radium". This occurs especially for γ-ray sources of higher activities and dimensions; when due to autoabsorption the sources cannot be regarded as point-sources.

According to the existing one point of view the concept of gRa is connected directly to the isotope itself. For any particular isotope one can determine the gRa value from the ionization constant Kγ of the isotope.

The other point of view attaches the gRa concept to any particular source of γ radiation. For this particular source the gRa value is determined by measuring its dose-rate in r/h at 1 m distance.

For γ-ray sources of higher activities and dimensions the gRa values calculated by the two methods can differ considerably.

Therefore there is a need to clarify the opinions about the gRa concept so as to be able definitively to formulate this radiologic unit.

Besides it is advisable to determine standard measuring methods of the gRa unit.

Sometimes, instead of the c or gRa values, one gives the "rhm" value (r/h at 1 m distance from the source). This value, pertaining to the source itself, is equivalent to the second concept of gRa. However, it does not give immediate information as to the activity of the source, which for many years has always been compared with radium standards.

While presenting all the above mentioned aspects of the problem the authors propose, that after clarifying of misunderstandings, it shall be recommended to use the gRa unit for radioisotopic γ-ray sources as used in industrial radiography and teletherapy.

La notion d' "équivalent-gramme de radium" pour les émetteurs γ non ponctuels

La notion d'équivalent-gramme de radium (gRa) est très utile en radiographie industrielle et en téléthérapie pour exprimer l'activité d'un émetteur γ. Cette notion, tout en donnant une idée de l'activité de la source, permet en même temps de déterminer les doses en roentgens/h auxquelles on peut s'attendre à une distance quelconque de la source.

Il existe une divergence d'opinion sur la notion même d'équivalent-gramme de radium. Cette divergence se manifeste surtout pour les émetteurs γ de haute
activité et de grandes dimensions, lorsque, en raison de l’autoabsorption, elles ne peuvent pas être considérées comme ponctuelles.

Selon l’un des points de vue actuels, la notion de gRa est liée directement à l’isotope lui-même. Pour tout isotope, on peut déterminer la valeur du gRa à partir du facteur d’ionisation Ky de l’isotope considéré.

L’autre conception rattache la notion de gRa à l’émetteur γ. La valeur du gRa d’une source est déterminée en mesurant l’intensité de la dose qu’elle émet en reoqntgens/h à un mètre de distance.

Pour les sources γ de haute activité et de grandes dimensions, les valeurs du gRa calculées selon les deux méthodes peuvent présenter des différences considérables.

Il y a donc lieu d’élucider la notion de gRa afin de pouvoir donner de cette unité radiologique une formulation précise.

Il est en outre souhaitable de fixer des méthodes normalisées de mesure de l’unité gRa.

Parfois, au lieu de valeurs en c ou en gRa, on donne la valeur en « rhm » (roentgens/h à un mètre de la source). Cette valeur, qui dépend de la source elle-même, équivaut à la deuxième conception du gRa. Toutefois, elle ne donne pas de renseignements immédiats sur l’activité de la source, qui, pendant des années, a toujours été comparée avec des étalons de radium.

Tout en présentant l’ensemble des aspects ci-dessus du problème, les auteurs proposent qu’après avoir mis fin aux malentendus, il soit recommandé d’employer l’unité gRa pour les émetteurs γ de la même manière qu’en radiographie industrielle et en téléthérapie.

Понятие «грамм-эквивалент радия» для неточечных изотопных источников β-лучей

Весьма полезно использовать понятие «грамм-эквивалент радия» (гРа) в промышленной радиографии и телерадиотерапии для обозначения активности изотопного источника γ-лучей. Давая представление об активности источника, это понятие позволяет в то же время определить мощность дозы в рентгенах в час (р/ч), которая предполагается на том или ином расстоянии от источника.

Существует расхождение во мнениях относительно самого понятия «грамм-эквивалент радия». Это относится особенно к источникам γ-лучей высоких активностей и размеров, когда из-за автоабсорбции эти источники не могут рассматриваться как точечные.

Согласно одной из существующих точек зрения, понятие гРа непосредственно связано с самим изотопом. Для каждого отдельного изотопа можно вывести величину гРа из коэффициента ионизации Ky определенного изотопа.

Другая точка зрения приписывает понятие гРа каждому отдельному источнику γ-радиации. Для данного отдельного источника величина гРа определяется путем измерения ее мощности дозы в р/ч на расстоянии одного метра от источника.

Для источников γ-лучей высоких активностей и размеров величины гРа, определенные с помощью этих двух методов, могут быть весьма различными.
В связи с этим необходимо выяснить точки зрения относительно понятия гРа с тем, чтобы можно было ясно определить эту радиологическую единицу.

Кроме того, целесообразно определить методы стандартного измерения единицы гРа.

Иногда вместо величины с или величины гРа дается величина дозы в один рентген в час на расстоянии одного метра от источника излучения. Эта величина, относящаяся к самому источнику, равнозначна второму понятию гРа. Однако она не дает непосредственных сведений об активности источника, которая в течение многих лет всегда сравнивается с радиевыми эталонами.

Представляя все изложенные выше аспекты этой проблемы, авторы предлагают после выяснения всех противоречивых точек зрения рекомендовать использовать единицу гРа для радиоизотопных источников γ-лучей, как они используются в промышленной радиографии и телетерапии.

El concepto de “equivalente gramo del radio” en las fuentes isotópicas de rayos γ, no puntiformes

En radiografía industrial y en teleterapia conviene utilizar el concepto de equivalente gramo del radio (gRa) para expresar la actividad de una fuente isotópica emisora de rayos γ. Este concepto da una idea de la actividad de la fuente y permite al mismo tiempo determinar la intensidad de la dosis en roentgens/hora a una distancia dada de la fuente.

Existen discrepancias de opinión acerca del concepto mismo de equivalente gramo del radio, especialmente en el caso de fuentes de rayos γ de grandes dimensiones y elevada actividad, cuando, debido a la autoabsorción, no pueden ser consideradas como puntiformes.

Según uno de los criterios existentes, el concepto de gRa está directamente relacionado con el isótopo. El valor gRa para un isótopo dado puede determinarse a partir de su constante de ionización $K_γ$.

Con arreglo al otro criterio, el concepto de gRa está relacionado con la fuente concreta de rayos γ de que se trate. El valor gRa se establece para una fuente dada midiendo la intensidad de dosis en roentgens/hora a un metro de distancia.

Los valores gRa, calculados según esos dos métodos para las fuentes de rayos γ de gran tamaño y actividad pueden diferir considerablemente.

Por ello, es preciso aclarar las opiniones sobre el concepto de gRa a fin de poder establecer con carácter definitivo esta unidad radiológica.

Además, es aconsejable fijar métodos uniformes de medición de la unidad gRa.

A veces, en lugar de expresar las actividades en curies o en gRa, se indican en “rhm” (roentgens/hora a un metro de distancia de la fuente). Este valor, que se refiere exclusivamente a la fuente, es equivalente al concepto de gRa citado en segundo lugar. Sin embargo, no da una noción directa de la actividad de la fuente, que durante muchos años ha debido referirse a patrones de radio.

Después de exponer todos estos aspectos del problema, los autores proponen que, una vez aclaradas las falsas interpretaciones, se recomiende el uso de la unidad gRa para las fuentes isotópicas de rayos γ, siguiendo la práctica adoptada en radiografía industrial y en teleterapia.

407
Activity, which for radiation sources is a basic concept, has no immediate meaning for radiation sources applied to radiography and teletherapy. In both cases, when using a source one wants to know the dose-rate of $\gamma$ radiation at given distance from the source. (Besides this value one wants to know also such physical constants of the source as $\gamma$-ray energy, half-period and geometrical dimensions.)

The effect which the $\gamma$ radiation evokes on the X-ray film (its blackening) in case of radiography or biological changes in tissues of the irradiated patient in teletherapy are both dependent on the radiation dose. So when the dose-rate (e.g. in r/h) is known one can immediately determine the dose for a given irradiation time.

The measurement of dose-rates of $\gamma$ radiation from the radioisotopes is neither a new nor a difficult problem. Measuring instruments available to-day permit to perform such measurements in a comparatively simple and speedy way. It is however quite impractical and impossible in the routine work to measure dose-rates for a given source each time before taking a radiographe or performing an irradiation of a patient.

Therefore it is necessary to specify for each radiation source such a radiation constant, from which it will be possible to determine the dose-rate at a given distance directly without the necessity to perform measurements or complicated calculations.

Unfortunately, when the source activity is given in curies it is impossible to determine directly the dose rate at a given distance from the source. It is, of course, possible to calculate the dose-rates from the activity in curies. To do that one must know both the disintegration scheme of the isotope in question as well as the ionization constants for all energy lines of the $\gamma$-ray spectrum.
When the γ-ray spectrum of the isotope is known as well as the relative intensities of each energy line, then one can calculate the dose-rates from the given curie value by referring to the well known diagram, shown in figure 1. The partial ionization constants for each energy line of the isotope and the total ionization constant $K_\gamma$ (expressed in r/ch at 1 m) can be found in appropriate tables, as it is rather inconvenient to calculate the total ionization constant from the above graph. There are also suitable equations for $K_\gamma$.

In many cases however the data pertaining to the radiation spectrum of the given radioisotope and its relative spectral lines intensities are divergent. (Here the Ir$^{192}$, which has for a long time been in practical use, can serve as an example.)

For high activity sources this problem becomes still more complicated. Then the dimensions of the source increase and the source can no longer be looked at as a point source. The autoabsorption in the source can no longer be neglected.

Of course in this case one can calculate also the interesting dose-rate taking into account the autoabsorption in the source. To do that one must know the absorption coefficient of γ-rays in the source itself, its dimensions and the exact location of the point at which the dose-rate has to be determined. Besides one must remember that for non-monochromatic γ radiation (and it is so with most of the radioisotopes) the absorption coefficient has not a constant value as it changes along the path of γ-rays. It is true that also in the case of our calculations one can use suitable equations and tables giving both absorption coefficients for different depths of irradiated material and permitting to calculate autoabsorption of γ-rays in the source itself. It is evident that all these calculations form a new barrier for the quick calculation of dose-rates for a given radiation source.

As can be seen it is rather a complicated and tiresome way to calculate dose-rates from the known source activity in curies. This is the more so for large radiation sources which nowadays are frequently used both in radiography and teletherapy.

Of course one can avoid all those difficulties by measuring directly the dose-rate at a given distance (usually 1 m) from the radiation source and giving this value as a specific constant of the source. This is often done in practice. One specifies the so-called “rhm” value, which gives the dose-rate in r/h at 1 m distance from the source.

The introduction of the “rhm” unit to designate the radiation source settles in principle the whole problem, for knowing the “rhm” value one can calculate directly from the inverse squares law the dose-rate at any desired distance from the source (if this distance is great enough in comparison to the source dimensions). The “rhm” value gives at the same time a certain idea about the source activity (not specifying it exactly).

In spite of all the above mentioned advantages the use of the “rhm” unit has this principal disadvantage that it gives not immediately the source activity, whereas it is useful to know the total activity of the source. Knowing it one can immediately judge the amount of radioactive substance present and see the possibilities and dangers caused by the source and its uses.

The historic development of applications of radioactive sources has introduced radium as a standard of γ radiation. Therefore from the very beginning one has always compared the activities of all other γ-ray sources with that of
radium. So when comparing the activity of any radiation source with that of radium one has the best idea about its strength.

For the reasons mentioned above it seems to be most suitable to express the activities of $\gamma$-ray sources for radiography and teletherapy in gramme-equivalent-of-radium — $gRa$ — units.

As it is well known the $gRa$ value of a given source can be determined by comparison of dose-rates of the source in question with those of a standard radium source. The comparison occurs by direct measurements.

For radium the gramme-equivalent-of-radium and curie are equivalent. On the other hand a radium source of a 1 c activity delivers a dose-rate of 0.84 r/h at 1 m, i.e. 0.84 rhm (the $\gamma$-rays of radium being filtered by 0.5 mm of Pt). So one has to remember only this value (0.84) and then for an activity given in $gRa$ one can calculate directly dose-rates in required distances.

The big advantage of specifying the activities of radiation sources in $gRa$ can be seen from the fact that it is easy to determine dose-rates of radiation and at the same time to be aware of the total activity of the source. Thus for practical applications the “$gRa$” unit is most convenient in use and is most figurative.

To use the “$gRa$” unit in practice one must first clarify the doubts which can arise about the concept itself.

According to one point of view the concept of “$gRa$” is connected directly to the isotope itself. For any particular isotope one can determine the $gRa$ value from the ionization constant $K_\gamma$, characteristic for the isotope, and its activity in c. In many tables one can find the $gRa/c$ values, calculated in such a way. Using those data one can determine the $gRa$ value for a given radiation source.

However, when calculating dose-rates (in r/h) from $gRa$ values and comparing the results of these calculations with direct measurements of dose-rates one sees that there is a difference due to the autoabsorption in the source, which was already stressed before.

Therefore, the other concept of $gRa$ gives another way to its determination. As the theoretical data of $gRa/c$ can in practice be used only for point-like sources the $gRa$ value shall be determined according to the measurements of dose-rates for a given, practical $\gamma$-ray source.

After measuring the dose-rate (and so the rhm value too) for a given source and distance one can calculate its $gRa$ taking into account that for 1 $gRa$ one has 0.84 rhm. The $gRa$ value determined by that method can be used now directly to dose-rate calculations.

The main disadvantages of this method for activity determination is the fact, that for radiation sources whose shape is no longer spherical the $gRa$ value depends on the direction from which the measurements were made. Those differences are more remarkable for smaller ratios of measuring distance and source dimensions.

Determining the $gRa$ value by the above method one can have an idea as to the total activity of the source but cannot determine it exactly in curies.

The authors are of the opinion that for radiography and teletherapy it is most convenient to specify source activity in $gRa$. One must, however, clarify the above mentioned differences of opinion as to the concept of $gRa$ itself. After doing so one ought to recommend to use the $gRa$ unit in practice and for all $\gamma$-ray sources for radiography and teletherapy specify always their activity

410
in gRa (which does not exclude the possibility of giving at the same time the activity in curies).

The next important problem connected with the introduction of the gRa concept is the problem of standard measurements. It is well known that dose measurements are largely dependent on γ-ray energies and the radioisotopes can have energies lower than 100 keV and higher than 2 MeV.

To illustrate quantitative differences which can be caused by the different interpretations of the “gRa” concept and lack of standard measuring methods one can cite the example of a Co\(^{60}\) radiographic source delivered by the Atomic Energy of Canada Ltd. to the Electrotechnical Institute in Warsaw (where both authors are employed). This source, when delivered, showed an activity of 715 c. As shown by the source certificate it is a cylindrical source and has a diameter of 15 mm and 15 mm height. The dose-rate measured by AECL at 1 m was 780 r/h.

Taking a 1.57 gRa/c value, as shown in various tables, one can calculate the gram-equivalent-of-radium as 930 gRa. If, on the contrary, one takes the Ky for Co\(^{60}\) as 1.32 r/ch at 1 m the gRa value will reach 1125 gRa. The difference is about 20%.

From the latter value (as calculated by the first method mentioned in the text) and taking into account the autoabsorption (calculated from the formula:

\[
\frac{a}{a_0} = 1 - e^{-\mu l} = 0.722
\]

where \(\mu = 0.46\) cm\(^{-1}\) for Co\(^{60}\) and \(l = 1.5\) cm) one obtains the dose-rate at 1 m as 684 r/h. This is about 15% lower as given by the AECL measurements giving 780 r/h.

This example shows clearly that there is an urgent need to define precisely the concept of gRa itself and specify standard measuring methods.
IV. 2. BATTERYLESS $\gamma$-RAY DOSIMETER

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Abstract

The Compton current produced by X-rays and $\gamma$-rays in the 0.3 to 3 MeV range is proportional to the radiant energy flux. Therefore the intensity of the current can be used as a measure of radiation dose. The present paper describes a radiation receiver and electrical measuring system suitable for Compton current measurements and results obtained with this device under irradiation from a 2000 curie Co$^{60}$ source.

Dosimètre de rayons $\gamma$, sans batteries

Le courant de Compton produit par des rayons X et $\gamma$ dans le domaine d'énergies compris entre 0,3 et 3 MeV est proportionnel au flux d'énergie rayonnante. C'est pourquoi l'intensité du courant peut servir de mesure de la dose de rayonnements. Le mémoire décrit un ensemble, appareil récepteur de rayonnements et appareil de mesure électrique, permettant de mesurer le courant de Compton et il expose les résultats obtenus, au moyen de ce dispositif, sous irradiation par une source de Co$^{60}$ de 2000 curies.

$\gamma$-дозиметр без батареи

Комптоновский поток, образуемый рентгеновскими лучами и $\gamma$-лучами в пределах от 0,3 до 3 мэв, пропорционален потоку лучистой энергии. Поэтому интенсивность потока может быть использована для измерения дозы радиации. В настоящем документе описываются радиационный приемник и электрическая измерительная система, пригодные для измерений с помощью комптоновского потока, и результаты, полученные с помощью этого устройства в ходе облучения источником Co$^{60}$ мощностью от 2000 кюри.

Dosímetro a rayos $\gamma$ sin batería

La corriente de Compton producida por rayos X y rayos $\gamma$ de energía comprendida entre 0,3 y 3 MeV es proporcional al flujo de energía radiante. Por consiguiente, la intensidad de la corriente puede servir para medir la dosis de radiación. En este trabajo se describen un receptor de radiaciones y un sistema de medición eléctrica apropiados para determinar la intensidad de la corriente de Compton, así como los resultados obtenidos mediante este dispositivo cuando se irradiía con una fuente de Co$^{60}$ de 2000 curies.

Principle of Operation

The Compton electrons produced in a scatterer by a beam of penetrating photons move preferentially in the direction of the primary photon beam. The flux of fast electrons constitutes a current, the intensity of which is proportional to the dose-rate of the radiation. The current can be measured if a dielectric is used as a scatterer. Thus a new electrical $\gamma$-ray dosimeter is obtained which needs no auxiliary voltage source.
A schematic diagram of such a dosimeter is shown in figure 1. Here 1 is a grounded top electrode covering the surface of a highly insulating and radiation resistant dielectric, 2 and 3 the measuring electrode; 4 is an insulating base plate. The top electrode is a thin sheet of metal or a layer of conducting paint and therefore does not produce a measurable absorption of the photon beam. The measuring electrode serves at the same time as electrode and absorber; it is made of a piece of metal of high atomic number and thick enough to absorb the photon beam almost completely. It prevents the primary radiation from reaching the base plate. The thickness of the dielectric is a multiple of the maximum Compton electron range.

The device operates as follows: the photon beam impinges on the top electrode, penetrates with little absorption the dielectric, and finally is absorbed in the measuring electrode. The dielectric separating the two electrodes acts as a scatterer for the production of Compton electrons. These electrons move essentially in the same direction as the primary photons, thus giving an electric conduction current which, in the dielectric, flows from the top electrode to the measuring electrode. If the measuring electrode is kept at ground potential (like the top electrode), a short circuit current flows in the system; if it is kept in open circuit, a negative voltage builds up. The intensity of the short circuit current or the slope of the voltage curve against time in open circuit gives the radiation dose-rate; the total voltage is a measure for the dose itself. The system is actually a \( \gamma \)-ray battery which directly transforms radiant into electrical energy.

The current in the dielectric

It was mentioned that the thickness of the dielectric between the two electrodes considerably exceeds the maximum electron range. Under these conditions it would be wrong to consider the measuring electrode as a collector electrode for the Compton electrons because only a small fraction of the electrons produced in the dielectric actually reaches the electrode. Let us suppose that \( D \) is the thickness of the dielectric, \( d \) the average electron range, and \( n \) the average number of electrons produced per second over one electron range. A charge \( e \) moving over the distance \( d \) anywhere inside the dielectric produces on the measuring electrode a charge \( e \frac{d}{D} \). The \( n \) electrons produced in one
“layer”, \(d\), therefore give a current \(n \cdot e \cdot d/D\). But the total number of layers is \(d/D\); therefore, the total number of electrons produced in the dielectric per second is \(N = n \cdot D/d\) and the total current is \(n \cdot e\). The number of electrons striking the measuring electrode is again only \(n\). Thus while the measuring current reflects the contribution from all electrons produced within the dielectric, the number of electrons reaching the electrode represents only a small fraction of the total electrons produced. This arrangement considerably reduces the influence of interface effects due to electron back-scattering, which otherwise would appear at the metal surface and distort the result.

A certain fraction of the primary beam might still penetrate the measuring electrode and reach the insulating base plate. There it can produce an electron current that flows to ground and therefore has a direction contrary to that of the electron current in the top dielectric which is flowing from ground to measuring electrode. This reverse current must be kept as small as possible. This can be achieved by preventing the primary radiation from penetrating the measuring electrode. For this reason the measuring electrode must also play the rôle of a radiation absorber.

The space charge in the dielectric

The measurable current is smaller than the Compton electron current due to space charge effects connected with the absorption of the photon beam in the dielectric. If there were no absorption the number of electrons produced anywhere within the dielectric would equal the number of electrons stopped, and no space charge or polarization effects could result. However, the photon beam undergoes absorption in the dielectric. Therefore, the number of electrons produced decreases with increasing depth in the dielectric. Accordingly more electrons are stopped and trapped in each volume element inside the dielectric than are produced. A negative space charge builds up which, due to the high resistance of the dielectric, has a very high relaxation time. The increase of space charge field produces a displacement or polarization current which again is contrary to the Compton electron current and thus reduces the measuring current. Since the absorption of the photon beam is small it can be expected that the polarization current is much smaller than the conduction current. A precise calculation entirely confirms this assumption. With very high radiation doses an additional effect might appear. The space charge field strength might become so high that it reduces the average range of the Compton electrons which eventually have not enough energy to run against the space charge field. Under these conditions the conductivity of the dielectric will have to be increased to reduce the relaxation time for the decay of the space charge.

The “metallized” dielectric

It has already been pointed out that electron back-scattering at the metal surface is of minor importance. This and other surface effects can, however, be completely avoided by a construction in which there is no “radiological” interface between the dielectric and the measuring electrode. For this purpose the measuring electrode is made of two pieces, a block of lead or some other metal of high atomic number and a block made of a dielectric of low atomic number covered on all surfaces by a very thin film of conducting paint, e.g. aquadag. The dielectric should be the same as that used for the insulator and scatterer. It covers the block of lead. Such a construction is shown in figure 2.
A dielectric completely covered by a conducting layer is a Faraday cage. Any charge contained in it is found as surface charge on the conducting layer, independently of the spatial distribution of the charge within. Electrically, an insulator covered by a conducting layer behaves like a metal. Experiments with static or slowly varying fields or charge distributions would not allow one to distinguish between such a system and a true metal. Thus "pseudo-metals" of very low atomic number can be produced in this way.

If the conducting layer is sufficiently thin, no electron or photon back-scattering or reflection occurs at the interface. The incoming radiation, whether electronic or photonic, does not "see" the interface and enters the electrode system without distortion. Electron and photon back-scattering eventually occurs inside the electrode system at the lead surface. The back-scattered radiation has, however, a much lower energy than the direct radiation and thus can be absorbed relatively easily still in the "metallized" dielectric.

Construction of radiation receivers

Figure 2 shows a non-portable radiation receiver. The measuring electrode system is formed by a lead block of 12 X 12 cm area and 12 cm height covered by a "metallized" block of plexiglas of the same area and a height of 2.5 cm. This system is completely surrounded by a plexiglas insulator of 2.5 cm thickness, the whole surface of which is painted with aquadag and grounded. The measuring electrode is connected to a suitable electrical measuring circuit through a shielded coaxial cable. Measurements were made by the Townsend compensation method in which the potential difference between measuring electrode and external electrode is practically kept at zero. In this way the
system operated in short circuit conditions and effects due to natural or induced conductivity and residual charges were minimized.

A portable receiver is shown in figure 3. It contains a simple measuring electrode of lead of about 1 cm thickness and $10 \times 10$ cm area contained in a plastic insulator covered by a conductive coating and a thin housing of metal foils. Access to the measuring electrode system is made by removing the insulating plug that normally closes the system. Measurements are taken after irradiation by means of a portable electrometer.

**Sensitivity**

Measurements were made with the receiver of figure 2 placed at a distance of 1 m from a Co$^{60}$ source of 2250 curie and a beam area of $8 \times 8$ cm. The calculated energy flux was $7 \times 10^6$ erg/sec. The measured current output was $8.8 \times 10^{-11}$ amp. This gives $1.25 \times 10^{-16}$ amp per erg/sec. The portable device can give a sensitivity of 0.5 volt per roentgen. A more detailed account of these measurements is given in a previous paper (1).

**Theory**

A detailed theory of the behaviour of the instrument has been developed and will be published in a forthcoming paper. For the sensitivity of the receiver mentioned above, the theory gives a value which, within the precision of the measurement, is in agreement with the experimental result. The theory also shows that the sensitivity increases slowly with increasing energy. If the sensitivity for the Co$^{60}$ radiation of 1.25 MeV arbitrarily is taken as 1, the sensitivity at 500 kV is 0.8 and at 2.5 MeV it is 1.1.

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IV. 3. CALORIMETRIC MEASUREMENTS OF γ-RAYS AND CALIBRATION OF FERROUS SULPHATE RADIATION DOSIMETER

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Abstract

An absolute determination of the high dose-rate in the 1200 curie Co⁶⁰ facility of the Danish Atomic Energy Commission has been performed by calorimetric methods. Experimental equipment and results will be discussed.

Mesures calorimétriques du rayonnement γ

Une détermination absolue de la dose émise par la source de 1200 c de Co⁶⁰ de la Commission danoise de l'énergie atomique a été obtenue par des méthodes calorimétriques. Le mémoire décrit le matériel expérimental utilisé et les résultats obtenus.

Калориметрические измерения γ-радиации

Абсолютное определение дозы высокой мощности в установке Датской комиссии по атомной энергии, где используется кобальт-60 на 1200 кюри, было осуществлено с помощью калориметрических методов. Будут рассмотрены экспериментальное оборудование и полученные результаты.

Mediciones calorimétricas de la radiación γ

Mediante técnicas calorimétricas se han llevado a cabo determinaciones absolutas de la intensidad de la radiación emitida por la fuente de Co⁶⁰ de 1200 curies perteneciente a la Comisión de Energía Atómica de Dinamarca. El autor describe el equipo experimental utilizado y los resultados obtenidos.

Introduction

Ferrous sulphate solution has in the recent years come into wide use as a dosimeter and as a secondary standard for γ dosimeters.

As a measure for the radiation dose one uses the yield of oxidation in a solution, which is $10^{-3}$ molar FeSO₄(NH₄)₂SO₄, 6 H₂O and 0.8 n H₂SO₄. NaCl is added up to $10^{-3}$ molar (to avoid disturbance by organic matter). The water is triple distilled (first distilled from chromic acid — then from potassium permanganate and, finally, purely distilled).

The oxidation yield may be measured by a spectrophotometer; one measures the absorption due to the Fe³⁺ ions at 3050 Å.

The measurements are reproducible within less than 0.4% when care is taken of the purity of the water and the other chemical components and of the temperature of the solution during the spectrophotometric measurements.

The yield is independent of γ- and electron-dose-rates between 0.1 rad/sec to $10^{10}$ rads/sec (1) (2), and it is independent of ion densities obtained by electron tracks (3).

It is therefore of great interest to know the G-value for the yield of oxidation in the Fe²⁺/Fe³⁺⁺ system (G represents the number of processes per 100 eV).
Until now the determination of the G-value has been limited by the accuracy of the absolute measurements of the dose. At the moment G-values (for γ- and electron-rays) between 15.3 and 15.8 are mainly used. The values most used are those measured by HOCHANADEL and GHORMLY \(^{(4)}\) who obtained \(G = 15.6 \pm 0.3\), and LAZZO, DEWHURST, and MILTON BURTON \(^{(5)}\) who obtained \(G = 15.8 \pm 0.3\). Their values are obtained by Co\(^{60}\)-γ-rays. SCHULER and ALLEN \(^{(1)}\) report \(G = 15.45 \pm 0.11\) obtained by cathode rays.

In the present experiment we have obtained a G value of 15.63 \(\pm\) 0.07 with a Co\(^{60}\) source. We used a molar extinction coefficient for the Fe\(^{+++}\) ions of 2196 \(\pm\) 6 mol\(^{-1}\) cm\(^{-1}\) at 25° C.

The Calorimeter

We have used a thermos flask in a geometry shown in figure 1 (a description of the irradiation facility is given in an earlier paper \(^{(6)}\).

![Ice-Calorimeter](image)

There are two Co\(^{60}\)-sources of 300 curies each on either side of a PVC jacket of 100 mm in diameter. The outside diameter of the thermos flask is 70 mm, the inside diameter being 60 mm. The volume of the thermos flask is 470 cm\(^3\). The thermos flask is made of glass. There is vacuum between the two mantles which are silver-coated on the vacuum side. The glass walls are approximately 1.0 mm thick.

The energy absorbed in the internal system of the thermos flask is measured by means of a capillary tube. The thermos flask is filled with ice and water, and the capillary tube is filled with water (all precautions are taken to avoid the presence of air in the system).

The volume of the ice and water decreases by an amount of 1.137 mm\(^3\) per calorie absorbed. By means of the capillary tube we measure the volume changes. A feature of this method of caloric measurements is that we do not introduce any thermometers that might disturb the measurements and that the volume changes give exactly the integrated energy absorption through the whole sample. Furthermore, the boundaries (the vacuum between the two mantles of the thermos flask) are well defined. Due to the fact that we have ice and water between the thermos flask and the PVC jacket, the external and
internal systems are symmetric at the boundaries. Due to the ice-water system used, the temperature outside and inside the thermos flask is self-regulating (the temperature outside the PVC jacket is kept very near 0° C).

When the average radiation field in the water and ice in the internal system is $X$ rad/hour per gram of water we get:

$$X (Q + q \cdot a \cdot b). 2,388 \cdot 10^{-6} = \frac{V_i}{1,137}$$

where "Q" is the mean amount of ice and water in grams; "q" is the weight of the inside part of the thermos flask; "a" is a correction factor for the different radiation intensity in the ice-water part of the interior and in the inner part of the thermos flask; "b" corrects for the different specific absorption per gram of glass and water. The factor $2,388 \cdot 10^{-6}$ is the energy of one rad in calories. $V_i$ is the volume changes in mm$^3$ per hour, and the factor 1,137 is defined above.

**Experiment I** (with water and ice in the PVC jacket)
- $Q = 445,34 \pm 0,30$
- $q = 64,00 \pm 2,00$
- $a = 0,92 \pm 0,01$
- $b = 0,893 \pm 0,01$
- $V_i = 186,89 \pm 0,96$

From equation (1) we then get

$$X_i = 138,358 \pm 0,085 \text{ rad/hour}$$

**Experiment II** (without ice water in the PVC jacket)
- $Q = 439,72 \pm 0,30$
- $q = 64,00 \pm 2,00$
- $a = 0,92 \pm 0,01$
- $b = 0,393 \pm 0,01$
- $V_i = 195,650 \pm 0,90$

From equation (1) we then get

$$X_{ii} = 146,396 \pm 0,080 \text{ rad/hour}$$

In order to get the corresponding energy absorption values per cm$^3$ of the ferro sulphate solution we must multiply these values by the specific gravity 1,0269. We may further multiply by 0.996 due to the fact that the field is reduced more in the heavier ferro sulphate solution than in the ice-water. Further, the factor 0.996 corrects for different specific absorptions of ferro sulphate solution and water.

The energy absorption $X_i$ per cm$^3$ in the ferro sulphate solution is then

$$X_i^1 = (138,358 \pm 0,085) \cdot 1,02693 \cdot 0,996 \cdot 0,996 = 140,947 \pm 0,085$$

$$X_{ii}^1 = (146,396 \pm 0,080) \cdot 1,02693 \cdot 0,996 \cdot 0,996 = 149,135 \pm 0,080.$$

The molar extinction coefficient at 3050 Å was found to be $2196 \pm 6 \text{ mol}^{-1} \text{cm}^{-1}$ at 25° C in accordance with the value found by SCHULER and ALLEN, 2174 ± 6 at 23.7° C \(^{(1)}\). The apparent difference between these values is due to the fact that the molar extinction coefficient changes approximately 15—18 per 1° C.

We measured the O. D. of $\frac{1}{3000}$ molar Fe$\text{SO}_4 \cdot (\text{NH}_4)_2\text{SO}_4$, 24 H$_2$O in 0.8 n H$_2$SO$_4$ solution, and it was found to be $0,732 \pm 2 \text{ cm}^{-1}$ at 25° C. Schuler and Allen measured instead on a ceri oxidized FeSO$_4$ solution. Our value differs by approximately 1% from that obtained by RIGG, TAYLOR, and WEISS \(^{(7)}\) and that used by Hochanadel. Rigg, Taylor and Weiss oxidized
FeSO₄ by H₂O₂. We also tried this method, but it did not give a well-defined value of the molar extinction coefficient, since the value found was somewhat dependent on the concentration of H₂O₂ used.

We get the G-values:

\[ G_1 = \frac{0.7534 \cdot 10^{-3} \cdot 6.022 \cdot 10^{28} \cdot 60}{2196 \cdot 9 \cdot 140.947 \cdot 6.254 \cdot 10^{14} \cdot 10^{-2}} = 15.65 \]

\[ G_2 = \frac{0.7945 \cdot 10^{-3} \cdot 60 \cdot 6.022 \cdot 10^{28}}{2196 \cdot 9 \cdot 149.135 \cdot 6.245 \cdot 10^{11}} = 15.60 \]

We have made the assumption that the total absorbed energy is converted to thermal energy. This might not be the case, since the absorbed energy could possibly also appear as:

1. Dissociation of the water molecules
2. Light emission
3. Crystal energy in the ice.

However, due to the fact that the ice is at the melting point we cannot expect any conversion to crystal energy.

Furthermore, all emission of light in the internal system will be absorbed in it due to the fact that the silver-coating of the inner mantle makes our system an optically closed system.

The problem of the dissociation of the water is somewhat more difficult to handle. The energy absorbed in this form must correspond to the G-value of H₂-production by irradiation. This G-value is approximately 0.4 when the H₂ is removed (9) corresponding to a correction of our G-value of 1.2% giving 15.45 instead of 15.63 obtained above. But when H₂ is not removed as in our system we have a back reaction which results in a much lower G-value for the dissociation of the water-ice system or a G-value of approximately 3% of the 0.4-value mentioned above. The steady state should occur after concentration of H₂O₂ less than \(10^{18}\) molecules per liter (5). This corresponds to approximately the same concentration of H₂, which means that in our case we should expect a steady state after less than half an hour of irradiation (the ice-water system was made up by distilled water and was almost free of air). Due to this fact we expect no correction from the dissociation of the ice-water system.

Our results are, therefore, \(G = 15.62 \pm 0.07\). The molar extinction coefficient of Fe⁺⁺⁺ used was 2196 ± 6 mol⁻¹·cm⁻¹ at 25°C. The optical density was measured by a ZEISS PMQ II spectrophotometer.

REFERENCES

DISCUSSION

Professor Aglintsev (Union of Soviet Socialist Republics) (Translated from Russian):

Mr. Chairman, I should like to make some comments on the question raised in Mr. Domanus' paper. The question we are faced with is whether or not there should be a radium standard. It is not very easy to abandon the radium standard, but it is equally difficult to give reasons for keeping it. If we weigh it up, we find that it will have many disadvantages — a complicated arrangement, the presence of a filter, costliness and, most important, the complicated nature of the radium spectrum in which close to 60 lines have already been found. Of course, the concept of a cobalt equivalent is simpler, and that of a caesium equivalent simpler still, than that of a radium equivalent. There is a certain merit in the radium standard. For metrologists of the older school it is a reminder of their youth, when radium was the sole important material, while for their younger colleagues it represents a certain glamour surrounding radium measurements. But the radium standard does have one great substantial advantage which makes it impossible for us to abandon it, and that is that there are very few isotopes from which standards can be prepared by weighing. They may be standards with a half-life of hundreds, thousands or even tens of thousands of years, and if we survey all our elements we find that by weighing we can prepare a radium standard or a plutonium standard. The latter fact may perhaps one day mean that we can create a curie standard based on a weighed quantity of plutonium, and by so doing take an important step away from measurement methods involving standards which, as Dr. Weiss has told us, are not free from elusive systematic errors, and also away from the problems of self-ionization in the preparation, and go over to direct weighing.

This, then, is the main reason for using the radium standard — it can be prepared by the weighing method. You cannot make a curie standard out of it for this is a very complicated system.

Now to the heart of the matter. You can say, if you like, that in metrology radium is necessary for just one thing — for the figure 0.825 — and that in essence radium is a model substance which produces a certain dose at a certain distance, i.e., radium is equivalent to the röntgen/hour/metre (r/h/m). That is its direct importance. Now the problems raised in the paper by our Polish colleagues boil down to the fact that it is difficult to separate the characteristics of materials and the characteristics of made-up articles. The concept of the grammie-equivalent is unexceptionable for a point source. It is unexceptionable for the characteristics of an isotope, although it may be unnecessary. But it loses its additive property when we go on to large made-up articles or preparations. If we take a large cylinder with a grammie-equivalent of radium and add a second cylinder, we do not obtain two grammie-equivalents of radium and self-absorption, but if the additive property is lacking, things are bad and life is hard.

Mr. Domanus (Poland):

As I said in my paper, I would not insist on the concept of grammie-equivalent of radium being linked to radium directly. It can equally well be
the r/h/m value. What I wanted to point out was that the methods now used are inadequate, there are big differences as to the interpretation of the concept itself, and we need to choose one method to be able to determine activities of large radiation sources in which self-absorption cannot be neglected.

Dr. Trott (United Kingdom):

First, I think from experience in England we would certainly agree with Mr. Domanus on the difficulties of knowing what one has got when one buys a tele-caesium unit. The reasons are evident — the people who send it out do not know what they have given you. Certain figures are stated as curies which may be based on long steps in chemistry and are not related to 4π measurements. I do not think that one should try to relate nuclides which are not radium to radium. It was once suggested that a particular water-board in a certain part of England should measure the radioactivity in its water and specify it in strontium units! I suggest that to do this with radium and caesium and cobalt, just to give people an idea of the activity, is equally misleading, since the maximum permissible amount of radium in the body is 0.1 of a microgramme while that of radiocaesium is many microcuries, and from that point of view it would be meaningless to say what is equivalent. From the radiation point of view, one measures the dose rate.

Now it seems to me that there are two points. First, if one wants, one must accept the problems of absorption in the source and make a dose-rate measurement, and presumably the best way is to refer to it as, say, caesium equivalents or cobalt equivalents, defining what one means there, for instance, by saying that if this were a source with no self-absorption, then the dose rate is so much and this would be x curies. This at least tells you what isotope you are working with. Secondly, I would say that the manufacturers who send these sources out lead us all into difficulties by putting a number on a piece of paper and circulating it with no information. I think this is most misleading and that in all cases where a figure is stated the way in which it is arrived at should be fully indicated. This could save a great deal of trouble and anxiety on the part of the people who receive the sources.

However, I think that expressing them all in radium equivalents is going to lead to a very large number of difficulties; we on the radiological side like to do this for isotope sources only on rare occasions when surgeons seem particularly keen to think in terms of radium, but in all other cases we would try to avoid this.

Mr. Domanus:

I agree with the first part of Mr. Trott's statement, but as regards the second part, what would be a better solution of expressing activities in a not misleading way than in gramme-equivalents of radium or in r/h/m values?

Dr. Lecoin (France) (Translated from French):

I should like to observe that the milligramme equivalent of radium is a unit which has been in use since 1912 to define the quantities of certain natural radioisotopes which cannot be weighed. It has long been conventional to speak of milligramme equivalents of mesothorium or of actinium, and this definition was subsequently extended to cover various artificial radioisotopes. This defini-
tion of the milligramme equivalent applies only to one specific apparatus — in this case it was the large Curie-Debierne condenser — and in respect of a specific lead absorption. Where these conditions are not fulfilled, one cannot speak of a milligramme equivalent. The calibrations hold good only for a specific apparatus and absorption. Many calibrations have been performed to relate the milligramme equivalent of radium to the number of curies contained in it. Some data are therefore to be found in certain tables. I believe that in these circumstances it would be preferable to choose another name for the unit you are proposing, which is likely to lead to confusion.

Mr. Domanus (Translated from French):
I also said in presenting the paper that there were difficulties with standard measurements and that there was a need to formulate with care the method by which it is intended to measure the gramme-equivalent of radium. I fully agree that a different unit can be selected; the point is that one should be selected and then agreed to by all countries producing radioactive sources, because countries such as ours buy isotopes from one supplier which are specified solely in terms of gramme-equivalents of radium and we buy isotopes from another supplier which are specified solely in curies, and subsequent comparisons of the data reveal considerable discrepancies of up to 20%.

Dr. Lecoin (Translated from French):
I fully agree, but I should prefer that, for this new unit which is to be defined, radium filtered through either 5 mm or 1 cm of lead should be used instead of through 0.5 mm of platinum. The use of the penetrating radiation of radium only will greatly facilitate comparison and make measurements much more accurate and easy. I have had to deal with several similar cases and found that radiation determinations using unfiltered radium are very unreliable. I think that the radium you work with must have been filtered through something with more energy than 0.5 mm of platinum.

The Chairman (Dr. Putman, United Kingdom):
There is a question, from Dr. Manov addressed to Mr. Brynjolfsson.

Dr. Manov (United States of America):
As I understand it, the basis of your method of measurement of \( \gamma \) radiation is the release of heat by the absorption of the \( \gamma \) radiation in the ice-and-water mixture. Now, could you increase the sensitivity of your method several times by adding heavy metals inside the innermost Dewar flask to increase this \( \gamma \) absorption?

Mr. Brynjolfsson (Denmark):
I wanted an inner system of thermistors almost equivalent to the ferrosulphate solution and therefore I was not interested in putting heavy materials in the system; but I have thought of the possibility for other purposes, and I believe this would be possible.
V.
MICROCALORIMETRIC METHODS OF STANDARDIZATION

(SESSION 6, 16 OCTOBER 1959)
V. 1. LA MÉTHODE DE MICROCALORIMÉTRIE ADIABATIQUE À COMPENSATION AUTOMATIQUE — APPLICATION À LA COMPARAISON DES ÉTALONS DE RA

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Abstract


A description of the new automatic compensation adiabatic microcalorimetric equipment built at the Curie Laboratory of the Radium Institute.

Potentialities and accuracy of the method.

Application to the comparison of radium standards.

La Méthode de microcalorimétrie adiabatique à compensation automatique. Application à la comparaison des étalons de Ra.

Description de la nouvelle installation de microcalorimétrie adiabatique à compensation automatique construite au Laboratoire Curie de l’Institut du Radium.

Possibilités et précision de la méthode.

Application à la comparaison des étalons de radium.

Метод адиабатической микрокалориметрии при автоматическом уравновешивании — Применение метода при сравнении эталонов радия

Описание новой установки адиабатической микрокалориметрии при автоматическом уравновешивании, построенной в лаборатории Кюри Института радия.

Возможности и точность метода.

Применение метода при сравнении эталонов радия.

El método de microcalorimetria adiabática de compensación automática. Su aplicación a la comparación de patrones de radio.

Descripción de la nueva instalación de microcalorimetria adiabática de compensación automática construida en el Laboratorio Curie del Instituto del Radio de París.

Posibilidades y precisión del método.

Aplicación a la comparación de patrones de radio.

L’utilisation de la calorimétrie pour la détermination des quantités d’énergies mises en jeu par les désintégrations radioactives remonte aux débuts même de la radioactivité.

Etant données les faibles quantités de chaleur mises en jeu, une série de techniques nouvelles furent proposées — techniques que l’on désigne sous le terme de «micro-calorimétrie».

Parmi toutes ces méthodes, nous avons préféré celle qui fut proposée par Swietoslawski et Dorabialsk, la méthode de calorimétrie adiabatique.

Nous rappelerons ici brièvement le principe de cette méthode:

Les sources radioactives dont on veut mesurer le débit de chaleur sont placées dans un calorimètre hermétiquement clos dans les parois duquel sont
absorbés les rayonnements émis. Ce calorimètre, suspendu dans une enceinte métallique par des fils très fins, est en principe isolé au point de vue thermique: les échanges de chaleur sont très petits et deviennent pratiquement nuls lorsque le calorimètre et l’enceinte sont à des températures voisines (quelques millièmes de degré). La manipulation consiste donc essentiellement à suivre du plus près possible l’évolution thermique du calorimètre en annulant constamment la différence de température existant entre l’enceinte et le calorimètre. Dans ce but, l’enceinte est plongée dans un bain d’eau d’une centaine de litres dont la température peut être augmentée progressivement par injections de petites quantités d’eau chaude. Dans ces conditions, l’élévation de température du bain, mesurée par un thermomètre au centième ou au millième de degré, est égale à l’élévation de température du calorimètre.

La différence entre le bain et le calorimètre est décelée à chaque instant au moyen d’un couple thermo-électrique en cuivre constantan, relié à un galvanomètre à haute sensibilité permettant d’apprécier $10^{-4}$ degré. Le déplacement du spot du galvanomètre dans un sens ou dans l’autre indique par conséquent une surchauffe, soit du bain, soit du calorimètre: il s’agit de maintenir constamment le spot très près du zéro de l’échelle en agissant sur la température du bain.

Cette méthode est d’une grande souplesse d’emploi. La forme et la masse des calorimètres utilisés peuvent être exactement appropriées aux différents phénomènes que l’on désire étudier, et la masse calorimétrique peut en être déterminée avec une grande précision.

L’intervention des couples thermo-électriques se limite à la détection d’une très faible force électromotrice sans qu’interviennent la signification et la nature de cette force dans des milieux fortement ionisés.

De plus, cette méthode qui est utilisée au Laboratoire Curie depuis 1925, nous était parfaitement connue.

Mais la précision des mesures était limitée, jusqu’à maintenant, par le dispositif de compensation de chaleur qui exigeait l’intervention constante d’un opérateur entraîné. La durée des mesures était de ce fait restreinte et la précision...
était fonction de l'habileté de l'opérateur. Nous nous sommes donc proposés de rendre automatiques toutes les opérations de compensation: nous exposons ici le principe de cette réalisation et un certain nombre d'applications immédiates concernant la détermination des périodes radioactives et la comparaison des étalons de radium.

Le dispositif que nous avons réalisé au Laboratoire Curie, en collaboration avec Mr. J. Robert, est basé sur l'emploi d'une cellule photo-résistante placée près du zéro de l'échelle. En l'absence de toute compensation, le calorimètre contenant une source radioactive s'échauffe seul, le spot dérive donc plus au moins rapidement sur l'échelle suivant l'intensité de la source radioactive. Au moment où il entre dans le champ de la cellule, l'éclaircissement produit donne naissance à un courant de quelques microampères, suffisant pour faire basculer un microrelais; celui-ci fait basculer à son tour une série de relais à forte intensité mettant en marche une minuterie qui actionne l'ouverture d'une électrovanne pendant un temps rigoureusement déterminé: l'injection d'eau dans le bain doit provoquer une très légère surchauffe de façon que le spot retourne en arrière et quitte le champ de la cellule. L'appareil est alors prêt pour un nouveau cycle.

Ce dispositif simple en principe se complique par le fait de l'inertie du système. L'équilibre thermique du bain après chaque injection d'eau chaude demande, avant d'être réalisé, 10 ou 15 secondes au bout desquelles les couples sont le siège d'une f. e. m. Il faut ensuite compter sur l'inertie du cadre du galvanomètre qui ne commencera à tourner que 10 ou 15 secondes plus tard; or, pendant ces 20 ou 30 secondes, le spot continue à avancer et peut quitter le champ de la cellule qu'il actionnerait de nouveau à son retour, provoquant une surchauffe importante. Ce simple exemple montre un aspect des différents problèmes qui se sont posés.

Le temps d'ouverture de l'électro-vanne est de l'ordre de la seconde, et doit être réglé au centième de seconde près.

On obtient dans ces conditions une suite ininterrompue de cycles durant une à deux minutes et comportant alternativement surchauffe du calorimètre et surchauffe du bain, ce qui se traduit par une oscillation forcée du spot du galvanomètre de part et d’autre du zéro. La courbe figure 2 donne une idée du déplacement du spot sur l'échelle en fonction du temps.

![Figure 2](image)

Oscillations du galvanomètre. Les flèches indiquent le déclenchement de l'électro-vanne au cours de chaque cycle.

Le dispositif ainsi conçu peut fonctionner sans surveillance pendant un temps indéfini; nous avons été amené à le laisser marcher 48 heures consécutives et l'on n'est guère limité en ce sens que par l'étendue de l'échelle thermométrique.

**Causes d’erreurs possibles.** — A. Echanges de chaleur: Des échanges
de chaleur entre l’enceinte et le calorimètre peuvent se produire soit par rayonnement, soit par conductibilité dans les fils et dans l’air.

Si l’on admet au cours de chaque cycle des différences de température n’excédant pas un millième de degré (oscillations du spot de 4 à 5 mm d’amplitude), les échanges de chaleur restent extrêmement faibles. De plus, ils se produisent alternativement dans l’un ou l’autre sens: par un réglage judicieux de la position de la cellule sur l’échelle, on arrive à annuler tout échange de chaleur en donnant au spot une même élongation de part et d’autre du zéro, ou plus exactement en faisant en sorte que la surface totale du cycle (t, △T) soit nulle; l’expérience montre que l’on obtient ce résultat en plaçant la cellule à 2 ou 3 mm du zéro, dans le sens de dérive du spot (figure 2).

La précision reste limitée par la dérive spontanée du zéro qui peut atteindre parfois deux millimètres par heure. Tout se passe alors comme si la compensation donnait en moyenne une surchauffe dans l’un ou l’autre sens. Des mesures faites avec un zéro volontairement décalé ont montré que les échanges de chaleur sont encore négligeables pour des dérives de quelques millimètres par heure, en particulier lorsque les mesures portent sur des sources pour lesquelles le débit de chaleur est considérable. Néanmoins il est encore nécessaire de ramener le galvanomètre au zéro plusieurs fois dans la journée lorsque les sources étudiées sont faibles.

B. Les autres causes d’erreurs sont négligeables. On élimine en particulier toutes les corrections de calibrage de la colonne thermométrique en opérant toujours entre deux mêmes graduations de l’échelle. La comparaison de deux sources se ramène, dans ces conditions, non pas à la mesure des élévations de température horaires produites, mais à la mesure des temps mis par le thermomètre pour franchir un même intervalle.

La lecture du thermomètre est faite en un point déterminé du cycle, par exemple au moment précis où le spot déclenche l’électro-vanne. Le tracé de la courbe d’élévation de température en fonction du temps pendant un certain nombre de cycles, au début et à la fin des mesures, permet d’évaluer avec une grande précision l’instant où le mercure du thermomètre passe devant la graduation prise pour repère.

Résultats expérimentaux

Mesures de périodes. — Nous avons tout d’abord étudié la sensibilité de notre dispositif en mesurant la décroissance de la chaleur dégagée par une source de radon dans un calorimètre de plomb pesant environ 300 grammes. Une première source a été suivie pendant 21 jours, son intensité passant de 82,3 à 1,9 millicuries et l’élévation de température horaire variant parallèlement de 960 à 21,7 millièmes de degrés. Les points obtenus se plaçaient sur une courbe exponentielle avec des écarts inférieurs à 0,5%.

Cette expérience montrait qu’il devenait possible de mesurer avec ce dispositif des sources d’intensité très différente. Notre appareil se prêtait donc particulièrement bien aux mesures de périodes, et l’on pouvait espérer obtenir par calorimétrie une précision au moins égale à celle que donnent les chambres d’ionisation. Nous avons donc entrepris une mesure précise de la période du radon: une deuxième source fut suivie pendant 15 jours, son intensité passant de 263 à 17,3 millicuries et l’élévation de température horaire variant de 2,536 à 0,167 degrés. Cette nouvelle expérience nous a donné pour la période:
T = 3,825 ± 0,004 jours

valeur communément admise jusqu’à présent.

Cette précision de la méthode calorimétrique nouvelle nous a conduit à l’utiliser pour comparer différents étalons de radium préparés par Höngischmid. En 1934, ce dernier a préparé à partir du chlorure de radium dont il s’était servi pour la détermination de la masse atomique du radium, une vingtaine de tubes contenant de 10 à 100 milligrammes de Ra élément. La précision de la détermination des masses de chaque étalon n’est pas indiquée sur les certificats signés par Höngischmid, mais il a indiqué par ailleurs que la précision de chacune de ses pesées était de 0,02 milligrammes. Comme la détermination de la masse de radium exige trois pesées, la précision de la détermination est comprise entre 0,06 et 0,1 milligramme de Ra élément. La plupart de ces tubes de radium sont actuellement utilisés par différents pays comme étalons nationaux de radium et les intercomparaisons effectuées au moyen du rayonnement γ pénétrant extérieur ont confirmé l’excellence des pesées effectuées par Höngischmid, dans la limite de précision indiquée plus haut.

Mais les techniques de comparaison par le rayonnement γ extérieur ont fait de gros progrès ces dernières années, la précision des déterminations étant souvent supérieure à 1/1000. Malheureusement il est extrêmement difficile de se servir de ces mesures pour connaître la masse réelle de radium contenue dans chaque tube. (Défaut de saturation des chambres d’ionisation — auto-absorption du rayonnement γ dans les grains de sels de RaCl₂ qui n’ont pas une épaisseur constante — géométrie légèrement différente par suite de la liberté du sel de radium dans les tubes, etc.) Dans l’impossibilité de procéder à de nouvelles pesées (et il serait certainement très difficile d’atteindre la même précision qu’Höngischmid), il était naturel de songer à comparer les débits de chaleur de ces tubes, qui sont directement proportionnels aux masses des sels de radium. L’ensemble de ce radium a en effet été préalablement débarrassé du RaD — à une date parfaitement connue — et l’accumulation du dépôt actif à évolution lente est la même pour chacun des tubes. La question de la géométrie des tubes et de la répartition du sel dans le tube n’intervient pas dans les mesures calorimétriques, et la détermination de la période du radon a montré qu’il était possible de comparer avec précision des débits de chaleur dont le rapport était de 1/10.

Deux séries de comparaisons ont été effectuées. Dans la première, exécutée en 1956, nous nous sommes servis du même calorimètre de 300 g de plomb utilisé pour la détermination de la période du radon. Les comparaisons ont été effectuées sur les trois étalons Höngischmid présents au Laboratoire Curie: le 5430 (étalon international nouveau: 16,92 mg de Ra sur le certificat Höngischmid); 5422/(24,78 mg) et le 5438 (38,06 mg). La comparaison des débits de chaleur se fait par la comparaison des temps mis par la colonne thermométrique pour parcourir la même portion de l’échelle thermométrique.

A titre d’indication:

Echelle thermométrique utilisé: = 2°655
Temps correspondant au 5430: T = 11 h 49 min 10 s .

La concordance des temps au cours des différentes séries de mesures a toujours été de l’ordre de 1 min. La précision des comparaisons calorimétriques est donc au moins du même ordre que la précision des pesées.
Le tableau I indique les résultats obtenus dans la comparaison des trois étalons au cours de cette série de mesures.

<table>
<thead>
<tr>
<th>Rapport des temps</th>
<th>Rapport des poids</th>
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<tr>
<td>( \frac{5422}{5430} ) = 1,4605 ± 0,005</td>
<td>1,4645 ± 0,01</td>
</tr>
<tr>
<td>( \frac{5438}{5430} ) = 2,2362 ± 0,005</td>
<td>2,2494 ± 0,01</td>
</tr>
<tr>
<td>( \frac{5438}{5422} ) = 1,530 ± 0,005</td>
<td>1,535 ± 0,01</td>
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Une deuxième série de comparaisons fut ensuite effectuée en utilisant un calorimètre de plomb de 70 g, de forme légèrement différente. La concordance avec la première série de mesure est supérieure à 1/500.

Enfin, nous avons réalisé depuis cette époque une nouvelle installation comportant en particulier un bain d’une contenance de 300 litres (au lieu de 100 comme précédemment).

Avec cette installation nous procédons actuellement à de longues séries de comparaisons entre l’étalon n° 5438 et 5422. La concordance des temps au cours de ces différents essais est supérieure à 1/1000. Nous avons pu en outre déterminer directement la masse calorimétrique de chacun des étalons et il est possible d’espérer par ce procédé comparer les masses de radium contenues dans ces étalons avec une précision supérieure à la précision des pesées.
Mr. B. W. Robinson (United Kingdom):
Could I ask Dr. Lecoin about his calorimetric method? I should like to have a little more detail about the thermocouple used for the differential temperature measurement and also to know whether he had to take any precautions, such as observing the mean temperature of the surface of his calorimeter, or whether he just took an arbitrary point on the calorimeter to attach the thermocouple to. In our own experience of precise calorimetry at the National Physical Laboratory, we have found the uniformity of the surface of the calorimeter when working to high accuracy to be of very great importance in relation to the vacuum jacket.

Dr. Lecoin (France) (Translated from French):
This is a very difficult question that Mr. Robinson is asking. The situation is as if the detection of a temperature difference by a copper-constantan thermocouple, consisting of two very fine wires about 0.01 mm thick, was perfect so long as the heat flux produced by the radioisotope led to a rise in temperature of more than 50 millidegrees per hour. Under these conditions, the results are comparable within the limits of present accuracy, no matter where the thermoelectric couples are placed. However, when the heat flux corresponds to a rise in temperature of less than 50 millidegrees per hour, the accuracy abruptly falls. It is as if there was from then on a kind of indetermination for the temperature at the surface of the thermocouple.

Dr. Trott (United Kingdom):
Dr. Lecoin mentioned that he had used thermistors instead of thermocouples in some experiments. Could he say a little more about the problems he met?

Dr. Lecoin (Translated from French):
This is a very difficult question. I think that the use of thermocouples to compensate for the Peltier effect otherwise than by zero methods is very risky because, at the present time, nobody knows — and anyway the question has not been studied — what an electromotive force in a very highly ionized medium is. That is why I prefer the method I have suggested, because the thermocouple has no function but that of zero detector. In other methods, and this applies to thermistors too, the use of groups of thermocouples or thermistors in highly ionized media is likely to entail large systematic errors.
VI.

PHOTOMULTIPLIERS

(SESSION 6, 16 OCTOBER 1959)
VI. 1. ON THE MECHANISM OF FATIGUE EFFECTS IN PHOTOMULTIPLIERS

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Abstract

New data on the fatigue effects in photomultipliers have been collected. The conclusions appear to be valid for all tubes with caesium compounds present in the system. The fatigue effect takes place in all the dynodes and — very likely — also in the photocathode.

The magnitude and time dependence of the instabilities vary with dynode current and ambient temperature. The recovery characteristics appear to depend mainly on the structure of the system.

The theory given on the mechanism of the fatigue effect shows, that the long-term instabilities are very likely caused by changes of the work function of the emissive layers.

Remarks are given as to what extent the influence of the photomultiplier instabilities on calibrating methods can be taken into account.

Le mécanisme des effets de fatigue dans les photomultiplicateurs

Les auteurs ont obtenu de nouvelles données concernant les effets de fatigue dans les photomultiplicateurs. Les conclusions qu’ils en tirent apparaissent comme valables pour tous les tubes dans la construction desquels entrent des combinaisons du césium. Les effets de fatigue peuvent se produire dans toutes les dynodes et également dans la photocathode.

L’ampleur et la dépendance en fonction du temps des instabilités varient avec le courant entre les dynodes et la température ambiante. Les caractéristiques de récupération semblent dépendre en général de la structure du système.

La théorie du mécanisme des effets de fatigue montre que les instabilités lentes sont vraisemblablement produites par des modifications de la fonction de travail des couches émissives.

Le mémoire s’achève par quelques considérations sur la mesure dans laquelle il est possible de tenir compte, dans les méthodes d’étalonnage, de l’influence des instabilités des photomultiplicateurs.

Об устройстве для определения эффекта усталости в фотоумножителях

Собраны новые данные об эффекте усталости в фотоумножителях. Полученные выводы, кажется, действительны в отношении всех труб, в системе которых используются цезиевые соединения. Эффект усталости отмечается во всех динодах, а возможно, и в фотокатодах.

Размеры нестабильности и ее продолжительность различны для различных динодов и температур. Регенерационные свойства зависят в основном от структуры системы.

Приводимая теория о механике эффекта усталости показывает, что длительная нестабильность вероятнее всего вызывается изменением рабочих функций эмиссионных слоев.

Приводятся замечания относительно того, как можно учесть влияние, оказываемое нестабильностью фотоумножителей на методы калибрования.
Sobre el mecanismo de los efectos de la fatiga en los fotomultiplicadores

Los autores han resumido nuevos datos sobre los efectos de la fatiga en los fotomultiplicadores. Las conclusiones que de ellos se desprenden parecen ser válidas para todos los tubos que contengan compuestos de cesio. El efecto de fatiga se observa en todos los dinodos y, muy probablemente, también en el fotocátodo.

La magnitud de las inestabilidades y su distribución temporal varían en función de la intensidad de la corriente dinódica y de la temperatura ambiente. Por otra parte, las características de recuperación dependen principalmente de la estructura del circuito.

En su exposición teórica sobre el mecanismo de los efectos de la fatiga, los autores demuestran que las inestabilidades a largo plazo se deben con toda probabilidad a alteraciones de la función de trabajo en las capas emisoras.

Por último, formulan algunas observaciones sobre la medida en que la influencia de las inestabilidades de los fotomultiplicadores puede tenerse en cuenta al calcular los resultados de las operaciones de calibrado.

I. Characteristics of electron-emitting surfaces

Fatigue effects of secondary electron emitters have been reported by several authors since many years \(^{1}\) \(^{2}\) \(^{3}\); the resulting gain variations in photomultiplier tubes are the major difficulty in all measuring methods where the detected pulses sum up to anode currents of the order of a tenth of a \(\mu\)A. Details of the activation of the dynode surface cause the gain either to increase or to decrease as it will be explained later on.

Various experiments of different authors lead to the conclusion, that fatigue effects in photomultiplier tubes can be explained by changes of the work function of the electron emitting surfaces: Clean surfaces can adsorb a monomolecular layer of residual gas atoms within 100 seconds at gas pressures of the order of \(10^{-8}\) mm Hg. Electropositive ions adsorbed at the surface of metals reduce the work function \(\varphi\) for electrons. The opposite effect is caused by adsorption of oxygen e. g. because of its large affinity to electrons.

An increase in gain i. e. an increase of the current of secondary electrons \(i_D\) could be caused by a pure emission of the adsorbed atoms, if one considered that only low energy electrons impinge on the surface of the dynodes. The electrons in the multiplier are accelerated between the dynodes to energies of the order of a hundred eV (a possible effect on the photocathode will be discussed later on). In this range of energy the changes of \(i_D\) are mainly caused by the changes of the work function \(\varphi\) as it will be shown below.

The velocity distribution of the secondary electrons can be considered to be a Maxwellian. Therefore the current of secondary electrons can be expressed by the formula:

\[
i_D = \frac{N c}{2 \sqrt{\pi}} \alpha e^{-\frac{\varphi}{v}}
\]

\(N c\) = Number of electrons impinging on the dynode
\(\alpha\) = most probable velocity of secondary electrons
\(\varphi\) = work function of dynode surface
\(v\) = mean velocity of secondary electrons in volts
A change in the characteristics of the photomultiplier only may cause a change in the work function, the other parameters remaining constant. 

\[ \ln \Delta i_D = \frac{\Delta \varphi}{v} \]

As example a change \( \Delta \varphi \) of 0.5 V may occur on the dynode surface. If one assumes \( v = 10 \) V it would give a gain variation of 5%. Both assumptions seem to be realistic for the usual conditions in a photomultiplier. On the photocathode one might expect only smaller changes in the work-function, but here the energy of the electrons released is much smaller too. This leads again to variations in the sensitivity of the order of some percent.

The various multiplier types (4) have different fatigue characteristics depending on the activation and the structure of the dynodes as it was indicated above. A decrease as well as an increase of the overall amplification was reported to take place in tubes of different types. CATHEY's report (3) seems to give the most representative example for the dependence of dynode gain on the amount of surface activation. Figure 1 shows the relation between dynode gain and amount of surface caesium for a Cs-Sb-O (cesium-antimony) dynode as they are used for instance in the RCA 6655's and 5819's. If the dynode is cesiated to some point such as “A” shortly below the peak of the curve, a decomposition of the dynode surface in the form of a diffusion of caesium off the surface will cause a continuous decrease in the gain. If it is cesiated to some point such as “B”, the fatigue will cause the gain first to increase than to decrease. The decomposition of the surface takes place when electrons with energies of the order of about 100 eV, which is well above the Cs-Sb-O binding energy strike the dynode surface. The released caesium will diffuse off the surface and create an undercesiated layer. At the same time there is a steady rate of recondensation of the caesium on all surfaces in the tube. When the dynode currents in the tube are switched off and the tube is not operating any more, the recondensation lasts until equilibrium and the original surface conditions are reached. It is easy to understand that the recovery takes a longer time for multiplier types with a linear dynode box structure (like the 6292 for instance) than for the tubes with a circular one (like 6342 and 5819).
In the tubes, that have Ag-Mg dynodes like the Dumont 6292 all the surfaces will also be covered with a certain amount of caesium since caesium-compounds are contained in the photocathodes. Such tubes will therefore fatigue like the overcesiated Cs-Sb-O dynodes until only a small amount of caesium is left.

II. Experimental set-up

Experiments have been carried out at the "Institut für Radiumforschung", Vienna, to investigate the fatigue-effects at different stages of the dynode structure of photomultipliers. The theoretical considerations show, that there might be any combination of equal or different gain-variation and, eventually, also variations of opposite sign at different points. This depends on the dynode-surfaces and the influence of caesium-condensation at the photocathode.

For the investigation a conventional scintillation spectrometer has been used, consisting of linear amplifier, single channel analyser, scaler, count-rate-meter, precision-high-voltage supply and a special preamplifier (Figure 2).

The typical stability, measured by the position of the Cs$^{137}$ photopeak is within $\pm 0.5\%$ per day. A weak Cs$^{137}$ source has been used for the gain-measurements, a stronger one (about 30 $\mu$C) for the irradiation. The intensity of the irradiation, or better, of the load on the photomultiplier was determined by measuring the current at the 10th dynode. The special preamplifier in use allows to measure the output pulse of the 6th and 10th dynode respectively and has also a sensitive, electrostatically shielded, microammeter for the d.c output current.

The loading of the photomultiplier was carried out at currents between 0.1 and 5 $\mu$A over intervals between 1 and 100 minutes. The gain variation was determined by measuring the Cs$^{137}$ photopeak of the weak source before and after the irradiation, the recovery by repeated measurements of the same kind (Figure 3). This procedure avoids any influence of overload.
on the electronic equipment, the gain is always determined under the same conditions.

III. Results

One RCA 6342 and four Dumont 6292 phototubes have been investigated, all of them showing fatigue effects. Different settings of multiplier gain (high voltage) and irradiation intensity giving the same load-current caused nearly the same effects. It was not possible to establish a definite relationship to these parameters.

The RCA tube showed a decrease of gain, the Dumont tubes an increase of different magnitude. The effect at the 6th dynode is in all cases smaller by a factor of about 5, for one Dumont tube the gain at the 6th dynode decreases while it increases at the 10th dynode. Figures 4 and 5 show the gain variations vs. irradiation time and collected charge respectively at the 10th dynode. There is some evidence for a dependence of the fatigue on the collected charge, but even at the same time there are different magnitudes of the effect for the same collected charge and different currents. In general the change is stronger at lower currents giving the same time-integral of charge. There is a saturation at high doses. The experiments did not give unique results about the recovery time. The RCA tube with circular dynode structure has short recovery-times between ½ and 2 hours, the Dumont tubes need about 10 to 20 hours to recover.

IV. Conclusions

In addition to the well-known fatigue effects at the normal output of a photomultiplier (10th dynode-anode) there is unquestionably an effect at the
lower stages. The gain variation at the 6th dynode is smaller than at the 10th dynode by a factor of about 5, but the pulse heights at the same points are different by a factor of 100. From there it follows that there is a gain variation at the 6th dynode, caused only by the load on the later stages, as the load on the earlier stages would cause only negligible effects (this has been in-

![Figure 4](image1)

![Figure 5](image2)

vestigated too). The fact of simultaneous gain-increase at the 10th dynode and decrease at the 6th of one tube gives again evidence for changes of, probably, the photocathode characteristics, caused by the load on the last dynodes. This is in a good agreement with the assumption that the electrons evaporate caesium from the dynode-surface (mainly at the last dynodes). As the concentration of caesium increases in the volume of the tube the condensation of the caesium on all surfaces increases too and might change especially the characteristics of photocathode. This effect should have no threshold of load current, because each electron has a certain chance to push a caesium atom out of the dynode surface. Only if the load current is low enough, that
recovery takes place faster than fatigue, the gain variations may be negligible compared to the instability of the electronic equipment. According to that it should be impossible to use 14-stage multipliers with high output currents (as usual in fast coincidence set-ups) simultaneously for precision spectroscopy even when taking the linear output of earlier stages.

The fatigue effects start at load currents well below 0.1 μA, i.e., at counting rates corresponding to 1000 pulses per second at an output level of some tenths of a volt at the photomultiplier output conditions which are quite common. For every kind of precision pulse-height measurements with a scintillation spectrometer it is necessary to check the tubes in use for their fatigue characteristics and the counting rates permissible in connexion with the high-voltage applied, the particle-energy and the scintillator characteristics. This may be done without difficulties, as load doses as high as 20 μA min. causing gain-variations up to 20% do not cause any remaining change in tube-characteristics.

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DISCUSSION

There was no discussion.
VII.

APPLICATION OF ABSOLUTE MEASUREMENT METHODS TO VARIOUS PROBLEMS

(SESSION 6, 16 OCTOBER 1959)
VII. 1. METHODS OF PARTICLE DETECTION IN FREE NEUTRON DECAY

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Abstract

A number of experimental studies have recently been completed by the Argonne Group on the decay of polarized neutrons in order to elucidate the structure of the weak nuclear interaction. These studies have taken the form of the measurement of the angular distributions of electrons and protons with respect to the spin direction of the decaying free neutrons. The basic components of the apparatus which will be discussed are:

1. The one-meter iron-cobalt mirror which selects a beam of highly polarized neutrons and the methods for determination of the polarization.
2. The electron detector comprising a 10 cm diameter, 6 mm thick mosaic of anthracene crystals, and its light pipe system.
3. The proton detector, a 14-stage electron multiplier system, the first stage with a $15 \times 15$ cm opening, tapering in four stages to a standard 10-stage multiplier structure, and its entrance baffles for angular resolution.
4. The electronic system which selects pulses from the detectors having the proper time sequence, relative time-delay and pulse-height to allow identification of a neutron decay.

Méthode de détection des particules dans une désintégration de neutrons libres

Le groupe Argonne a terminé récemment des études expérimentales sur la désintégration des neutrons polarisés, en vue de mettre en lumière le processus de l’interaction nucléaire faible. Ces études consistaient à mesurer les distributions angulaires d’électrons et de protons par rapport à la direction du spin des neutrons libres en voie de désintégration. Les principaux éléments de l’appareillage qui sera décrit sont:

1. Le miroir de ferro-cobalt d’un mètre, qui isole un faisceau de neutrons hautement polarisés, ainsi que les méthodes permettant de déterminer la polarisation.
2. Le détecteur électronique qui comprend une mosaïque de 10 cm de diamètre et de 6 mm d’épaisseur en cristaux d’anthracène, et son système sélectif.
3. Le détecteur de protons, un système multiplicateur électronique à 14 étages, le premier ayant une ouverture de $15 \times 15$ cm se rétrécissant en 4 étages pour aboutir à un multiplicateur ordinaire à 10 étages et ses chicanes d’entrée pour résolution angulaire.
4. Le système électronique qui isole dans les détecteurs les impulsions venant des détecteurs et ayant les caractéristiques voulues de fréquence, de retard relatif et d’amplitude et permet ainsi d’identifier la désintégration d’un neutron.

Метод обнаружения частиц при распаде свободного нейтрона

Недавно Аргоннной группой был завершен ряд экспериментальных исследований по распаду поляризованных нейтронов с тем, чтобы пролить
свет на структуру слабого ядерного взаимодействия. Эти исследования носили форму измерения угловых распределений электронов и протонов по отношению к направлению спина распадающихся свободных нейтронов. Основными узлами прибора, который будет обсуждаться в докладе, являются:

1. Железо-кобальтовое зеркало размером в 1 метр, собирающее пучок высоко поляризованных нейтронов, и методы определения поляризации.
2. Электронный детектор диаметром в 10 см с 6 мм мозаикой антраценовых кристаллов и системой осветительных трубок.
3. Протоновый детектор с системой электронного множителя в 14 каскадов, причем первый каскад с зазором 15 X 15 см; детектор собирается в 4 каскада до стандартного 10-ти каскадного множителя; вход его отводит угловое разложение.
4. Электронная система, отбирающая импульсы у детекторов и имеющая соответствующую последовательность во времени, относительную задержку времени и импульсную высоту для определения распада нейтронов.

Método para la detección de partículas en la desintegración de neutrones libres

En el Laboratorio de Argonne se ha llevado a cabo recientemente una serie de estudios experimentales sobre la desintegración de neutrones polarizados, con el fin de elucidar las modalidades de la interacción nuclear a bajas energías. Estos estudios han consistido en mediciones de la distribución angular de electrones y protones con respecto al sentido del spin de los neutrones libres que sufren la desintegración. Los componentes fundamentales del aparato que el autor describe en la memoria son:

1. Un espejo de hierro y cobalto, de un metro, que selecciona un haz de neutrones altamente polarizados y permite determinar el grado de polarización aplicando distintos procedimientos.
2. El detector de electrones, consistente en un mosaico de cristales de antraceno de 10 cm de diámetro y 6 mm de espesor, y en un dispositivo de canalización luminosa.
3. El detector de protones, formado por un sistema multiplicador de electrones de 14 etapas, la primera de las cuales con una abertura de 15 X 15 cm, que va disminuyendo en cuatro pasos hasta una estructura multiplicadora usual de 10 etapas, y sus deflectores de entrada para lograr la resolución angular.
4. El sistema electrónico que selecciona los impulsos que, al ser adecuados los intervalos con que se suceden, su defasaje en tiempo y su amplitud, hacen posible la identificación de una desintegración neutrónica.

The work which I would like to discuss briefly is a part of the experiments which were undertaken at Argonne National Laboratory as a co-operative experiment by Drs. M. Burgy, V. Krohn, R. Ringo and myself of Argonne, and Prof. V. L. Telegdi of the University of Chicago. These were a series of experiments designed to study the angular asymmetries of electrons and anti-neutrinos emitted in the simplest β decay of all, that of the free neutron. The angular asymmetries were measured with respect to the spin of the neutron. We used a beam of polarized neutrons. The results of these experiments (1) (2) have been
of assistance in determining the structure of the $\beta$ decay interaction including the invariances of the $\beta$ decay interaction under the transformations of parity, charge conjugation and time reversal. I will not go into these results here, but I propose to discuss some of the detectors which were developed in order to make the measurements.

The basic components of the experiment are first a cobalt mirror which one uses to obtain a beam of polarized neutrons, an electron detector, and a proton detector to detect the recoils from the neutron decay.

In a sense these experiments represent the essence of many years contact with the problems of $\beta$ counting, for here we have a beam of pure neutrons, a weightless source, indeed. We don't have to worry about self-absorption problems, finally, and the radiochemical purity is very high. In fact, you see, we not only have only neutrons, but only neutrons with their spins pointing in one direction. I must admit that we do have a slight impurity of photons in the neutron beam.

Well, let me just briefly sketch the principle of the cobalt mirror. The idea is that one makes a thin sheet of cobalt which is magnetized in a direction perpendicular to the direction of the incoming neutrons. We have a mirror made up of 5 inch squares, 10 of them, so that the whole mirror is about 50 inches long. The neutron beam impinges on this surface at an angle of 8 minutes. Neutrons are then reflected from the surface if their spin is in the right direction. This is a case where — due to the magnetic properties of cobalt for scattering of neutrons — one spin state will be critically reflected; the other spin state of the neutrons will not be reflected. Thus one obtains essentially a one hundred percent polarized beam of neutrons. The beam of neutrons which comes from a reactor has a distribution in energy, a Maxwellian distribution, with a peak at about .025 eV. Our mirror reflects all neutrons which are of

![Schematic drawing of neutron decay apparatus.](image-url)
lower energy than about $0.015 \text{ eV}$. This upper limit is determined by the angle to the beam at which the mirror is set. This angle is prescribed by the particular slit system which one has so that one does not transmit a direct beam through from the reactor. About $10\%$ of all the neutrons in the beam are reflected from the mirror and into our apparatus.

Figure 2
Top view of vacuum chamber.
Figure 1 shows a top view of the apparatus. The neutrons are collimated by slits one quarter inch wide and 85 inches apart. The mirror is about 50 inches long and it subtends about ½ inch of this beam when it is set at an angle of about 8 minutes to the beam. One can do very nice optics with thermal neutrons and select the reflected beams very easily and arrange shielding to prevent direct beam transmission. The neutrons that are reflected have their spins either pointing up or down into the plane of this paper, depending on the direction of the magnetic field across the mirror surface. The beam then goes through a thin copper window, a vacuum chamber which is about 6 ft long, 2 ft high and 1½ ft wide, through a slot in a lithium-lead alloy block which absorbs stray scattered neutrons, and then into the region in which we examine neutron decay. There is a sensitive region about 6 inches long from which we can detect particles which are emitted if neutrons decay on drifting through this region.

Figure 3
View into end of vacuum chamber. Electron asymmetry arrangement.

We have of the order of $5 \times 10^7$ neutrons per second, and when calculated in a distance of about 6 inches, this amounts to about 3 or 400 neutron decays per minute in this active region. A proton detector, with perhaps a collimator to determine the initial direction of proton emission when one wants to do angular measurements on the proton emission, and an electron detector which is a plastic, or an anthracene scintillator, are then located and combined in various
ways to determine the angular dependences of interest. The polarization of the beam itself is determined by reflecting again from another cobalt mirror at the exit end of the apparatus. If, for example, the beam is completely polarized and is reflected from another mirror that is magnetized in the same direction as the initial mirror, then all the neutrons will be reflected from the second mirror. Now, if you depolarize the neutrons, which is very easily done by simply putting a thin sheet of steel after the polarizing mirror (the magnetic domain are oriented at random and the neutron spins on going through from one domain to another are rapidly flipped over) so that at the second mirror half the neutron spins are in each direction and therefore only half will reflect. In this way, one can measure the polarization of the beam.

Figure 2 shows a top view of the vacuum chamber — just to give you an idea of the simplicity of the apparatus. The cobalt mirror is underneath a pile of concrete blocks. The neutron beam comes through a shield wall into the vacuum tank. The neutron beam is of the order of $\frac{3}{4}$ inch wide and about 5 inches high. One must have a guide field provided by magnetic condenser plates so that the neutrons do not precess in the earth's magnetic field. The proton counter looks in from the side, the electron detector is enclosed by sheets of iron which shield the photomultiplier from the guide field. All of this equipment was actually gathered together in a very short time as happens when experiments look very exciting.

Now then let me go to the individual detectors. Figure 3 shows a view into the end of the vacuum tank looking directly as the neutron beam. On the
right side is the electron detector — about 5 inches in diameter — inside of a lead shield. On the left can be seen the first stage of a proton detector which has an entrance opening 6 inches square. The dynodes are of silver magnesium alloy. The protons in this case — this is the arrangement for determining the electron asymmetry — are accelerated into the proton detector. In this case the neutron spins lie along the electron-proton detector axis and we want to see which end of the neutrons preferentially emit electrons. So you just count all of the protons you can, just simply to help identify electronically the neutron decays, and then measure the number of electrons emitted, say parallel with the neutron spin and then rotate the neutron spin in the opposite direction and see how many come out anti-parallel. Due to the non-conservation of parity we obtain different numbers and the actual value of this asymmetry is a very critical number for the theory of $\beta$ decay.

Figure 4 is a schematic drawing of this picture. The neutron beam is as indicated, most intense at the center with a height at half intensity of about 5 inches. There are two grids by means of which the protons are accelerated into the proton detector.

Figure 5 shows a picture of the proton detector. This is a Dumont type of structure. The first stage is $6 \times 6$ inches square and tapers down to about $3\frac{3}{4} \times 3\frac{3}{4}$ inches, the entrance size of the next stage. There are two more stages
of multiplication with successive tapering down of the aperture size. A standard 10 stage Dumont structure which has had the end of the tube removed is located at the exit aperture of the last stage so that the electrons can be collected into it and undergo ten more stages of multiplication. In this way we obtained a very large sensitive area for the proton detector.

The pulse distribution extends to very small pulse-heights and although very high efficiency can be obtained if one is willing to accept very high background, we chose an operating discrimination level at which we had 20—30% efficiency for proton detection. One ordinarily has single backgrounds of the order of 10 to 15,000 counts a minute when one is trying to measure the order of a few counts a minute due to neutron decay.

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Figures 6 and 7 give a picture of the anthracene detector crystal mosaic. The pieces of anthracene are 1 1/4 inch squares, and we obtained 16 of them. They were cut from one piece by the Harshaw Chemical Company and we checked to see that each one gave the same electron energy resolution and pulse height. They were cemented to a half-inch thick piece of plastic using a clear epoxy resin mixture. The edge was machined off so that we had a circular detector about 5 inches in diameter and about 3/4 inch thick.
The over-all resolution of this detector for electrons is of the order of 20% full width at half height for the cesium electron line. One can with a small crystal on a photomultiplier tube obtain about 12% resolution. This is the best that we have been able to do and only by using a British EMI 5 inch photomultiplier, at the end of a 6 inch long plastic light pipe. There may be other tubes available in the world which will also have a fairly uniform distribution of the photocathode sensitivity. The American tubes do not.

The last thing to say is how one determines, when one has observed a neutron decay. The protons require about a tenth of a microsecond to drift over to the proton detector. The electrons are collected in less than a millimicro-second. We use a sequence sensitive time-overlap coincidence circuit, which requires that a pulse comes from the proton detector later than a pulse from the electron detector. The pulses from the detectors are put into the time overlap circuit. The output from this circuit is proportional to the time delay between the two pulses. This distribution of pulse heights is displayed on a multichannel pulse-height analyzer and then one can pick out the neutron decays, because they are those counts which fall at a point corresponding to a tenth of a microsecond delay of the pulse from the proton detector. Prompt coincidences which are much more abundant than the neutron decays then appear at zero delay.

Figure 7
Anthracene mosaic detector. Top view.
and a random background is spread out underneath the whole distribution of pulses. The neutron decays give a peak several times background and by measuring our coincidence counting rates with the beam polarized, with the beam unpolarized, and with the various arrangements of the detectors, we can then make the required measurements on the neutron decays.

We are now in the process of refining our measurements in order to determine more exactly the ratio of vector to axial vector interactions in β decay. We are studying further the polarization of the neutron beam. The original results were obtained with about 90% polarization. We have at the moment, in one position of the mirror, been able to obtain 100% polarized neutrons. So we are confident that any uncertainties which we may have in the polarization will be resolved very soon and then we may continue on with the measurements.

The proton detector is also some source of trouble because the efficiency is not 100%. We detect only of the order of 20% of the protons which hit the detector. It would be very nice, of course, if we could increase this efficiency and obtain higher counting rates.

In conclusion let me list the major results of our experiments to date concerning the nature of the β decay interaction.

a) The parity and charge conjugation operations are not invariant operations in β decay.

b) The time reversal operation (or the product of parity and charge conjugation operations) is an invariant operation in β decay.

c) The vector and axial vector types of interaction predominate in β decay with a maximum contribution of about 30% of scalar and tensor type of coupling; S/V < 3, T/A < 3.

d) The vector and axial vector types of interaction contribute to the extent $A^2/V^2 = 1.55 \pm 1$ or $A/V = 1.25 \pm 0.05$.

e) The relative phase angle between the vector and axial vector interaction amplitudes is $180^\circ - 16^\circ$.

f) The anti-neutrino is right handed; the direction of spin lies along the direction of momentum.

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VII. 3. ESTIMATION OF A DISTRIBUTED $\beta$ EMITTER BY MEASUREMENTS OF BREMSSTRAHLUNG

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Abstract

In tracer experiments involving large, solid samples, the direct measurement of $\beta$ particles is restricted to those which originate near the surface of a sample. The greater penetration of bremsstrahlung, produced when $\beta$ particles are stopped in the body of the sample, allows contributions to be detected from a much larger volume.

$^{45}$Ca was used to measure the transfer of calcium from refractory to metal in a steel furnace, cast ingots containing the tracer isotope being used for measurement.

The $^{45}$Ca content of these ingots was estimated by comparison with a dummy ingot, built up to the same size and shape with laminated steel sheets. A thin lamina, carrying a measured quantity of $^{45}$Ca, was placed at various depths in the dummy ingot to obtain the counting rate (from bremsstrahlung) contributed by layers of the tracer at these depths and the resulting curve of counting rate versus depth was plotted. By integrating this curve, the total counting rate corresponding to a known uniform distribution of the tracer was deduced. The counting rates from sample ingots, measured under identical geometrical conditions, were then a good measure of their total calcium content. Calcium concentrations of a few parts per million were determined by these means.

Estimation des émetteurs $\beta$ non ponctuels par la mesure du rayonnement de freinage

Dans les expériences à l’aide d’indicateurs radioactifs portant sur de grands échantillons solides, on ne peut mesurer directement que les particules $\beta$ qui proviennent du voisinage de la surface de l’échantillon. La plus grande pénétration du rayonnement de freinage, qui se produit lorsque des particules $\beta$ sont arrêtées dans la masse de l’échantillon, permet de déceler les particules $\beta$ provenant d’un volume de matière beaucoup plus important.

Le $^{45}$Ca a servi à mesurer le transfert de calcium du revêtement réfractaire au métal dans un four sidérurgique, en utilisant pour la mesure des lingots contenant l’isotope radioactif.

La teneur en $^{45}$Ca de ces lingots a été estimée par comparaison avec un lingot d’essai fait de tôles d’acier laminées et ayant les mêmes taille et forme que les lingots de l’échantillon. Une plaque mince, portant une quantité connue de $^{45}$Ca, a été placée à différentes hauteurs dans le lingot d’essai; on a pu ainsi mesurer le taux de comptage (pour le rayonnement de freinage) dû à des couches de l’indicateur situées à ces hauteurs et en faire la représentation graphique en fonction de la hauteur. En intégrant cette courbe, on a calculé le taux global correspondant à une répartition uniforme connue de l’indicateur. Les taux de comptage pour les lingots de l’échantillon, mesurés dans des conditions géométriques identiques, ont donc fourni une mesure satisfaisante de la teneur totale en calcium. Des concentrations de calcium de l’ordre de quelques parties par million ont pu ainsi être décelées.
Подсчет распределенных β-излучателей путем измерения тормозного излучения

При опытах с меченными атомами, для которых требуются крупные, твердые образцы, прямое измерение β-частиц ограничено измерением тех β-частиц, которые образуются у поверхности образца. Большое проникновение тормозного излучения, получаемое тогда, когда распределение β-частиц приостановлено в образце, позволяет обнаружить его в гораздо большем объеме.

Са⁴⁵ был применен для измерения превращения кальция из огнеупорного материала в металл в сталой печи, причем для измерения применялись отлитые болванки, содержащие меченные атомы. Содержание Са⁴⁵ в этих болванках было вычислено путем сравнения с пробной болванкой, которая с помощью ламированных стальных листов была доведена до того же размера и формы. Тонкий ламированный слой, содержащий измеренное количество Са⁴⁵, был помещен на различной глубине в пробной болванке для получения скорости счета (тормозного излучения) от слоя с меченными атомами на этой глубине, и на карту нанесена полученная кривая скорости счета в зависимости от глубины. Путем интегрирования этой кривой была выведена полная скорость счета, соответствующая известному однородному распределению меченых атомов. Скорость счета, полученны из образцов болванок, измеренная при идентичных геометрических условиях, стала хорошим измерителем всего содержания кальция в них. Этим способом была определена концентрация кальция в нескольких частях на миллион.

 Evaluación de la actividad de un emisor β no puntiforme por medición de la radiación de frenado

En los trabajos con trazadores en que se utilizan muestras sólidas de grandes dimensiones, sólo pueden medirse directamente las partículas β que tienen su origen en las proximidades de la superficie de la muestra. El mayor poder de penetración de la radiación de frenado que se produce cuando las partículas β son frenadas en la masa de la muestra, permite detectar la actividad emitida por un volumen mucho mayor de la muestra.

Se ha utilizado Ca⁴⁵ para medir el paso del calcio del material refractario al metal en un horno siderúrgico. Para llevar a cabo las mediciones se utilizaron lingotes de colada que contenían el trazador.

El contenido de estos lingotes en Ca⁴⁵ se calculó por comparación con un lingote testigo, de idénticas dimensiones y forma, construido con placas de acero laminado. En el lingote testigo se colocó a diferentes profundidades una lámina delgada conteniendo una cantidad conocida de Ca⁴⁵; se determinó la velocidad de recuento (por medición de la radiación de frenado) correspondiente a la actividad emitida por las capas del trazador a dichas profundidades y los valores obtenidos se representaron gráficamente en función de la profundidad. Integrando la curva que así resulta se dedujo la velocidad global de recuento correspondiente a una distribución uniforme y conocida del trazador. Las velocidades de recuento de los lingotes tomados como muestra, determinadas en idénticas condiciones geométricas, proporcionan entonces una medida satisfactoria de su contenido total en calcio. De este modo se detectaron concentraciones de calcio de unas pocas partes por millón.
In a measurement of calcium retention cast iron, Ca\textsuperscript{45} was used as a tracer at a specific activity of 1.7 mc/g.

Samples for measurement were in the form of ingots of diameter 2.7 inches and thicknesses near to 2 inches. A sample of the initial calcium tracer was retained in solution as CaCl\textsubscript{2} for reference and comparison. Owing to the low maximum energy of the β particles from Ca\textsuperscript{45} (0.25 MeV) it was evident that only a shallow layer of calcium distributed in the ingot could be detected directly by its β particle emission.

However, measurements taken with an end-window β counter \textsuperscript{5}) on one flat face of the ingot showed that the counting rate obtained was very little reduced by the insertion of aluminium absorbers sufficient to stop the β particles completely. Furthermore the comparison of two samples, one of which had been diluted with inactive iron showed that the counting rate obtained was at least approximately proportional to the calcium content.

Examination of the source material revealed no penetrating radiation likely to account for the counting rates obtained. Figure 1 shows an absorption curve obtained with a 2 μc source made by evaporating the reference solution. Subsequent measurements after 4 months showed that the half-life of the observed radiation was compatible with of Ca\textsuperscript{45} (153 days).

\textsuperscript{5) Type PHM 2: General Electric Co., Ltd., Wembley.}

![Figure 1](image_url)

It was therefore suspected that the penetrating radiation was bremsstrahlung produced by the deceleration of Ca\textsuperscript{45} β particles throughout the body of a sample. In view of the small range of the β particles, the bremsstrahlung from such a thick sample might be expected to contribute more to
the counting rate than would the \( \beta \) particles originating from the surface layers, and being still proportional in intensity to the amount of \( \text{Ca}^{46} \) present would offer a convenient means of assay.

To verify this and to measure the tracer calcium in the samples a model was made from a series of inactive iron discs of different thicknesses, which could be built up to simulate the measured sample. A "uniform" layer of \( \text{Ca}^{46} \) was made by evaporation of 58 measured drops from a dilution of the aliquot solution regularly spaced over the surface of an iron disc 2.7 inches in diameter and 0.006 inch thick. This was covered with a shim steel disc of thickness 0.002 inch and was inserted at various positions in depth of the model. Measurements of counting rate were made with an end-window counter mounted above the block. These are plotted as a function of depth in figure 2, which shows the contribution of a known quantity of radioactive calcium at any depth in the block. The samples were also screened with 0.002 inch of shim steel and counting rates were measured under identical conditions for comparison.

![Figure 2](image)

To relate the results to the counting rate obtained from the calcium distributed through the samples, the following argument was used:

Let \( M \) be the total mass in grams of a sample of thickness \( D \) and let \( C \) be the mass of calcium per unit mass of iron it contains.

Then \( MC \) is the total mass of calcium in the sample.

Let \( m \) be the mass of calcium on the comparator disc, as used in the model, and \( f(x) \) the counting rate obtained when this is situated at depth \( x \).

Consider now an element of the unknown sample at depth \( x \) and of thickness \( dx \).
This contains \( \frac{MC}{D} \) dx grams of calcium.

Then the contribution to counting rate from this element is:

\[ \frac{MC}{mD} \int_0^{x=D+0.002} f(x) \, dx \]

And the total counting rate \( R \) is:

\[ R = \frac{MC}{mD} \int_0^{x=D+0.002} f(x) \, dx \]

Whence the concentration \( C \) is given by:

\[ C = \frac{mDR}{M \int_{0.002}^{x=D+0.002} f(x) \, dx} \]

The integral was determined by graphical integration of the curve in figure 2 and since the other parameters were measured, the value of \( C \) was deduced.

**Results**

Measurements were made on two castings, whose respective thicknesses were 1.925 inches and 1.75 inches. In making the second sample, the initial melt containing the tracer had been diluted by the addition of an equal quantity of inactive iron. The concentration \( C \) of labelled calcium in the second sample was therefore expected to be one half that in the first sample.

The results obtained were as follows:

<table>
<thead>
<tr>
<th>TABLE I</th>
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<tbody>
<tr>
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<tr>
<td>Calcium in comparator disc</td>
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<tr>
<td>Mass of sample</td>
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<tr>
<td>Thickness of sample</td>
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<tr>
<td>Counting rate from sample</td>
</tr>
<tr>
<td>Statistical deviation in ( D+0.002 ) f(x) dx (from Figure 2)</td>
</tr>
<tr>
<td>Grams (Ca)/Grams (Fe)</td>
</tr>
<tr>
<td>Estimated overall error (rms)</td>
</tr>
</tbody>
</table>

Samples identical in composition were analysed by radioactivation analysis for total calcium content by SMALES (1), together with an untreated sample containing no radioactivity which had been used in the above experiments for determinations of background counting rate. All samples were found to contain
from 5 to 6 parts per million of calcium, indicating that the original iron was already saturated with calcium at this level, but that the dissolved calcium exchanged freely with the large excess of added radioactive calcium (added at 500 parts per million).

Discussion

A criticism of the measurements just described is that the distribution of tracer within the ingots was not established and that local concentrations of calcium could have been present in the form of inclusions. This is considered unlikely, since measurements on opposite ends of samples yielded counting rates which were equal within the experimental accuracy, and also in view of the agreement of results with those obtained by radioactivation analysis.

It is established, in any case, that low energy $\beta$ emitters may still be used for tracer experiments involving samples of large mass, by measuring their bremsstrahlung, provided that the distribution in a sample is uniform, or at least known and reproducible. The activity per gram in sample 2 is approaching the lower limit which is conveniently measurable by these means in iron. This corresponded to $3.34 \times 10^{-6} \times 1.7 \text{ mc/g} = 0.0057 \mu\text{c/g}$ and the total activity in sample 2 was approximately $6.5 \mu\text{c}$.

Acknowledgements

It is a pleasure to record the co-operation of Messrs. Mond Nickel, who sponsored these experiments and prepared the metallurgical samples, and of Messrs. E. W. Solomon and R. A. Peart, who between them made most of the experimental measurements.

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DISCUSSION

Dr. Curran (United Kingdom):

In the very elegant equipment described by Dr. Novey it puzzled me to note that a multiplier with a special anode was used as the proton detector although the efficiency measured was 20%. Could not a scintillator have been used?

Dr. Novey (United States of America):

We did try to make a zinc sulphide detector, but were unsuccessful. We did not spend a long time on it, because we wanted to have something that would work. The multiplier worked; we did not succeed in making a scintillator very efficient.

Dr. Curran:

Daly at AWRE has developed a special detector which uses a secondary emission stage (the same voltage accelerates ions on to the emitter and secondary electrons from it) and a scintillating material as detector. This is looked at with a commercial photomultiplier and, with ions from a mass spectrometer (energy 6—16 keV), seems to be very efficient. The work is in course of publication and can be communicated to Dr. Novey after the session.

Dr. Trott (United Kingdom):

Can Dr. Novey say a little more about the problem of selecting 5” photomultipliers? His remarks on the relative quality of United States and British tubes are of interest in view of the discussion this morning.

Dr. Novey:

The American 5” tubes were made by Dumont and were selected out of twenty or so. The EMI tube was perhaps the best out of three that I think we had. You see, if you have a large light pipe and a large scintillator on the other end, which is as big as the sensitive area on the photo-tube, the light tends to end up in about the same position from which it started out. Therefore you do not really make the process more uniform by using a longer light pipe and it seems you rely on the uniformity of the cathode surface. A lot of the British tubes were considerably better, and it was fairly easy to select one which gave us this 20% resolution instead of perhaps 30 or 40% with the corresponding American tubes. I think it is a matter of care in the production lines. It is not that we cannot produce tubes; it is just that few companies, once they have an item in production, are prepared to make slight changes in their production lines unless there is some big demand for it. One can, I think, buy selected tubes at a higher price, but it is much simpler just to be able to take them off the shelf.

Dr. Allen (United Kingdom):

There is one question I would like to ask Dr. Novey. You mention that you only used zinc sulphide as a scintillator to attempt to detect the protons. A publication appeared fairly recently indicating that the nature of particles...
can be determined, from the decay time, in a phosphor. I wonder whether perhaps with yet another time-sorting device in your electronics you might use an ordinary scintillator and differentiate between the protons and the γ-ray background, instead of using a straight multiplier as you have done.

Dr. Novey:

Well, very simply you just do not have enough energy — there is only about 12 keV. That system works if you have energies of several hundreds of keV or of several MeV.

Mr. Forte (Italy):

Just a word, which may not even be necessary after Dr. Novey's answer. There are two different methods of discriminating scintillation pulses by pulse-length discrimination. One, by employing organic scintillators, may be useful for protons down to a limit of about 0.5 MeV. The other one has not been applied to technical purposes, I think, but may be useful even at lower levels when using caesium iodide or zinc sulphide, but I do not know what the energy limit is.

Mr. Gasstrom (International Atomic Energy Agency):

Could you not have accelerated the protons a little more — not initially, but in the second stage?

Dr. Novey:

When you begin to accelerate them more you begin to accelerate everything else that is left in the vacuum system and the background count goes way up.

**CONCLUSIONS**

The Chairman (Dr. W. Mann, ICRU):

As the representative of the International Commission of Radiological Units and Measurements (ICRU), it has fallen to my lot to draw the final conclusions of this Symposium. In my capacity as a scientist, however, I have learned that very few conclusions can be looked upon as final and that the conclusions of today serve merely to sustain us in the tasks of tomorrow. My remarks on this subject therefore will be most brief.

The International Commission of Radiological Units and Measurements has been pleased to co-sponsor with the Agency this Symposium on the Metrology of Radionuclides. It has, I think, been a very good symposium. How do we now look ahead? Presumably our task lies in achieving greater precision and greater accuracy in the measurement of radioactivity. I think we may say that for most medical, diagnostic and therapeutic purposes we have already achieved limits of error that are less than the variations in the response of most biological systems. For the hospital physicist, therefore, the need is for more standards having about the same accuracy as we are at present producing. For the nuclear physicist, however, we have not yet attained the degree of accuracy that is required. Mr. B. W. Robinson has commented on the difficulty of increasing the accuracy of radioactive metrology from the order of ± 1% to the order of ± 0.1%. It is interesting to reflect, however,
that if we were to measure one microcurie for one hour the precision of our counting experiment would only be of the order of 1 in $10^4$. This, then, represents approximately the limit of accuracy to which we can aspire, and we can scarcely hope to achieve the accuracies of, say, 1 in $10^9$, associated with some other branches of metrology and physics. Even if we are measuring curie instead of microcurie quantities of radioactivity, we are faced with most serious problems of geometry and self-absorption, and the measurements may in turn depend upon measurements in the microcurie range. It is, however, possible that the direct measurement of energy emission may lead to greater precision, if not accuracy, in the curie range. Dr. H. P. Robinson has already obtained accuracies in a counting of the order of a few parts in $10^5$, and the beautiful work of Mr. Campion and his colleagues at Chalk River has pointed the way to greater accuracy in the assay of $\beta$ and $\gamma$ emitters. In this connexion it is somewhat amusing to consider that even though the difference between the curie and the velocity of light is only about 1 cm., we still have a long way to go to achieve the kind of accuracy associated with the measurement of velocity of light.

Dr. Manov has mentioned the problem of low-level radioactive contamination of pure reagents. In this connexion I would like to say that the ICRU Committee I on Standards and Measurements of Radioactivity for Radiological Use has already set up a Sub-Committee, under the able chairmanship of Mr. Trott, to consider this problem. Mr. Grinberg is also a member of the Sub-Committee and we hope to enlist the help of two or three other distinguished workers in this field.

Lastly, I should perhaps briefly refer to the question of international standards. In general, in view of the short half-lives of many of the nuclides involved, it is necessary for the national standardizing laboratories to maintain national standards, often in the form of a standard instrument used in conjunction with a long-lived reference source. We believe that frequent inter-comparisons between the standards of the various national laboratories is desirable, and for about the last five years the ICRU has been attempting to correlate the results of such inter-comparisons. The International Bureau of Weights and Measures, which was established by international treaty long before even the League of Nations was conceived, has already had considerable experience in this kind of correlation between the work of the national laboratories, especially in the field of electrical standards. The ICRU therefore proposes to hand over its responsibility in the field of radioactivity standards in the matter of correlation to the International Bureau of Weights and Measures. The Agency is also entering the field of radioactive metrology and is setting up a standards laboratory here in Vienna, with the help of Dr. Gandy. I feel sure that all the national laboratories will welcome the opportunity of inter-comparing their standards with those of the Agency when they become available, and I have already offered the co-operation of the United States National Bureau of Standards in any way possible.

In conclusion I would like, on behalf of all of us, to congratulate the Agency, and especially Professor Sanielevici, on the outstanding success of this Symposium, and I wish to express our very deep appreciation for all that Professor Sanielevici has done to make it so successful. I would also like to
express our gratitude to Professor Sanielevici's staff, and also to Mr. John Cummins, Mr. de Mautort and their staffs, including our more remote friends, the hard-working interpreters, who, coping with such a variety of unusual subjects, have contributed so much to our deliberations. It might indeed be said that they have dwelt in glass houses and upon our every word.

Prof. Sanielevici (International Atomic Energy Agency) (Translated from French):

We have now reached the end of our work. The Agency thanks you for the concentrated work to which you have been subjected and we apologize for what we might call the nuclear density of this symposium. We thank Dr. Mann for the undeservedly kind remarks he has made about us, and we would like to ask him to convey our sincere gratitude to the International Commission of Radiological Units for their assistance in the organization of our Symposium — assistance which is proved by the presence of nearly all the members of the Subcommittee on Radioactivity of the ICRU and by their interesting contributions to our work. We are going to end this Symposium in the hope that in the near future we will be able to gather you together again for another symposium on a subject which is allied to that of the present one. This will be a symposium on Selected Topics in Radiation Dosimetry. We wanted to announce this new symposium immediately and, of course, will keep you informed of the details of the development of the project. Once more I thank you all, and, on behalf of the International Atomic Energy Agency, I declare this symposium at an end.
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